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(74) Agents: YAMAMOTO, Ryozo et al.; Torimoto Kogyo Building, 38, Kanda-Higashimatsushitacho, Chiyoda-ku, Tokyo 101 (JP).

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(71) Applicant *(for all designated States except US)*: ISHIHARA SANGYO KAISHA LTD. [JP/JP]; 3-15, Edobori 1-chome, Nishi-ku, Osaka-shi, Osaka 550 (JP).

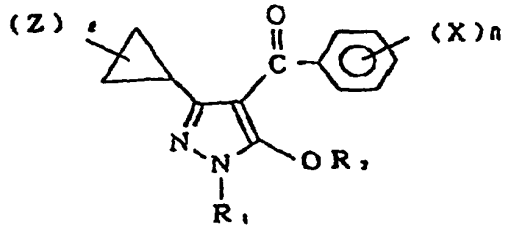
(72) Inventors; and

(75) Inventors/Applicants *(for US only)*: MURAI, Shigeo [JP/JP]; Ishihara Sangyo Kaisha Ltd. Chuo Kenkyusho, 3-1, Nishi-shibukawa 2-chome, Kusatsu-shi, Shiga 525 (JP). KIKUGAWA, Hiroshi [JP/JP]; Ishihara Sangyo Kaisha Ltd. Chuo Kenyusho, 3-1, Nishi-shibukawa 2-chome, Kusatsu-shi, Shiga 525 (JP). NAKAYAMA, Hitoshi [JP/JP]; Ishihara Sangyo Kaisha Ltd. Chuo Kenkyusho, 3-1, Nishi-shibukawa 2-chome, Kusatsu-shi, Shiga 525 (JP). SANO, Makiko [JP/JP]; Ishihara Sangyo Kaisha Ltd. Chuo Kenkyusho, 3-1, Nishi-shibukawa 2-chome, Kusatsu-shi, Shiga 525 (JP). ISOGAI, Akihiko [JP/JP]; Ishihara Sangyo Kaisha Ltd. Chuo Kenkyusho, 3-1, Nishi-shibukawa 2-chome, Kusatsu-shi, Shiga 525 (JP).

(54) Title: PYRAZOLE COMPOUNDS, PROCESSES FOR THEIR PRODUCTION AND HERBICIDES CONTAINING THEM

(57) Abstract

A pyrazole compound of formula (I) or its salt wherein R_1 is an alkyl group, R_2 is a hydrogen atom, a methyl group, -A-R_3, a phenyl group which may be substituted, a pyridyl group which may be substituted, or an allyl group which is substituted by a phenyl group, A is -SO_2-, -CO-, -$CH(R_6)$- or -$CH(R_7)$CO-, R_3 is an alkyl group which may be substituted, an alkenyl group which may be substituted, an alkynyl group which may be substituted, an alkoxy group which may be substituted, a cyano group, a dialkylamino group or a phenyl group which may be substituted, each of R_6 and R_7 is a hydrogen atom or an alkyl group, X is a hydrogen atom, a halogen atom, an alkyl group, a haloalkyl group, an alkoxy group, an alkylthio group, an alkylsulfinyl group, an alkylsulfonyl group, a nitro group, an alkoxycarbonyl group, -$SO_2N(R_8)R_9$, -$N(R_{10})SO_2R_{11}$, -$CH_2S(O)_qR_{12}$ or -OSO_2R_{13}, each of R_8, R_9, R_{10}, R_{11}, R_{12} and R_{13} is an alkyl group, Z is an alkyl group, l is an integer of from 0 to 5, n is an integer of from 1 to 5, and q is an integer of from 0 to 2, provided that when l is at least 2, a plurality of Z may be the same or different, and when n is at least 2, a plurality of X may be the same or different.



(I)

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DESCRIPTION

PYRAZOLE COMPOUNDS, PROCESSES FOR THEIR PRODUCTION AND HERBICIDES CONTAINING THEM

TECHNICAL FIELD

The present invention relates to novel pyrazole compounds useful as active ingredients for herbicides.

BACKGROUND ART

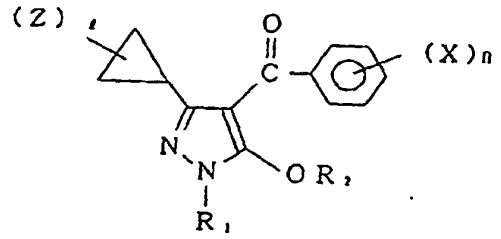
UK 2002375A and EP 282944A disclose pyrazole derivatives having various substituents at the 3-position of a pyrazole ring. However, the pyrazole compounds of the present invention are clearly distinguished from such derivatives in that they have a cycloalkyl group substituted at the 3-position of a pyrazole ring.

Further, EP 638555A discloses pyrazole glycolic acid amide derivatives having various substituents at the 3- and 4-positions of a pyrazole ring. However, the pyrazole compounds of the present invention are clearly distinguished from such derivatives in that they have a substituted benzoyl group substituted at the 4-position of a pyrazole ring.

DISCLOSURE OF THE INVENTION

The present inventors have conducted various studies paying attention to pyrazole compounds to find out an excellent herbicide and as a result, have accomplished the present invention. Namely, the present invention provides novel pyrazole compounds of the formula (I) or

their salts:



(I)

wherein R_1 is an alkyl group, R_2 is a hydrogen atom, a methyl group, $-A-R_3$, a phenyl group which may be substituted, a pyridyl group which may be substituted, or an allyl group which is substituted by a phenyl group, A is $-SO_2-$, $-CO-$, $-CH(R_6)-$ or $-CH(R_7)CO-$, R_3 is an alkyl group which may be substituted, an alkenyl group which may be substituted, an alkynyl group which may be substituted, an alkoxy group which may be substituted, a cyano group, a dialkylamino group or a phenyl group which may be substituted, each of R_6 and R_7 is a hydrogen atom or an alkyl group, X is a hydrogen atom, a halogen atom, an alkyl group, a haloalkyl group, an alkoxy group, an alkylthio group, an alkylsulfinyl group, an alkylsulfonyl group, a nitro group, an alkoxycarbonyl group, $-SO_2N(R_8)R_9$, $-N(R_{10})SO_2R_{11}$, $-CH_2S(O)qR_{12}$ or $-OSO_2R_{13}$, each of R_8, R_9, R_{10}, R_{11}, R_{12} and R_{13} is an alkyl group, Z is an alkyl group, l is an integer of from 0 to 5, n is an integer of from 1 to 5, and q is an integer of from 0 to 2, provided that when l is at least 2, a plurality of Z may be the same or different, and when n is at least 2, a

plurality of X may be the same or different; processes for their production; herbicides containing them; and novel intermediate compounds useful for producing them.

Now, the present invention will be described in detail with reference to the preferred embodiments.

The alkyl group or the alkyl moiety for R_1 and R_3 may be a C_{1-10}, preferably C_{1-5}, linear or branched alkyl group, and the alkyl group for R_6 and R_7 may be a C_{1-2} alkyl group. The alkyl group or the alkyl moiety for R_8, R_9, R_{10}, R_{11}, R_{12}, R_{13}, X and Z may be a C_{1-4} linear or branched alkyl group. Specific examples of such an alkyl group or moiety include methyl, ethyl, propyl, isopropyl, butyl, tert-butyl, pentyl, octyl and decyl. The alkenyl group for R_3 may be a C_{2-10} linear or branched alkenyl group, such as vinyl, allyl, butadienyl or isopropenyl. The alkynyl group for R_3 may be a C_{2-10} linear or branched alkynyl group, such as ethynyl, propynyl or 2-penten-4-ynyl.

The substituent for the phenyl group which may be substituted or the pyridyl group which may be substituted, for R_2, may be halogen, C_{1-4} haloalkyl or nitro. The number of substituents may be one or more, and when the number is at least 2, a plurality of such substituents may be the same or different.

The substituent for the alkyl which may be substituted, the alkenyl which may be substituted, the alkynyl which may be substituted, or the alkoxy group

which may be substituted, for R_3, may be halogen, C_{1-4} alkoxy, C_{1-6} alkoxycarbonyl or cyano. The number of substituents may be one or more, and if it is at least 2, a plurality of such substituents may be the same or different.

The substituent for the phenyl group which may be substituted, for R_3, may be halogen, C_{1-4} alkyl, C_{1-4} haloalkyl, C_{1-4} alkoxy-C_{1-4} alkyl, C_{1-4} alkoxy, nitro or cyano. The number of substituents may be one or more, and if it is at least 2, a plurality of such substituents may be the same or different.

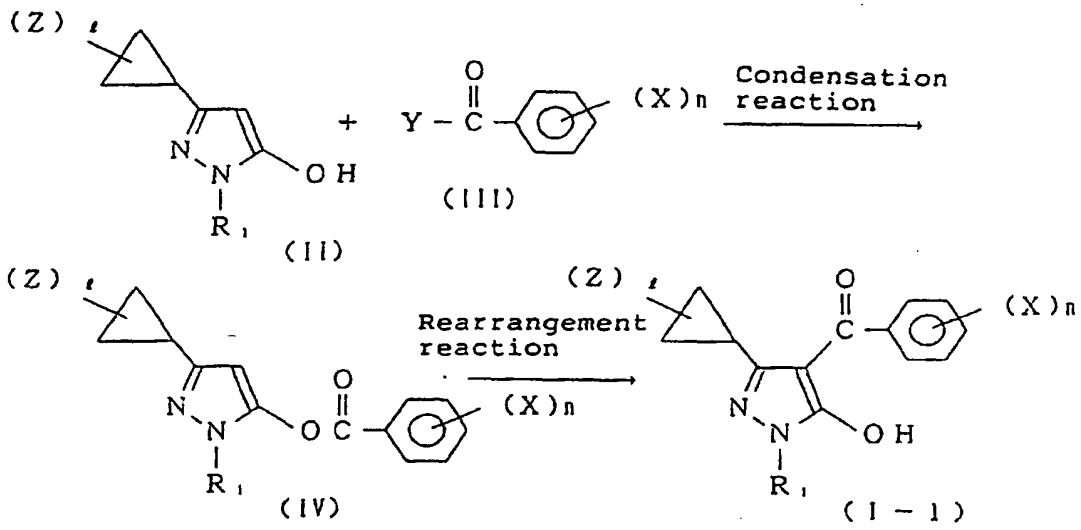
The halogen atom for X and the halogen as the substituent contained in R_2, R_3 and X, may be a fluorine atom, a chlorine atom, a bromine atom or an iodine atom. The number of halogen atoms as substituents, may be one or more, and if it is at least 2, a plurality of halogen atoms may be the same or different.

Among pyrazole compounds of the formula (I), a compound wherein R_2 is a hydrogen atom, is capable of forming a salt. The salt may be any salt so long as it is agriculturally acceptable, and it may, for example, be an alkali metal salt such as a sodium salt or a potassium salt, an alkaline earth metal salt such as a magnesium salt or a calcium salt, or an ammonium salt such as a dimethylamine salt or a triethylamine salt.

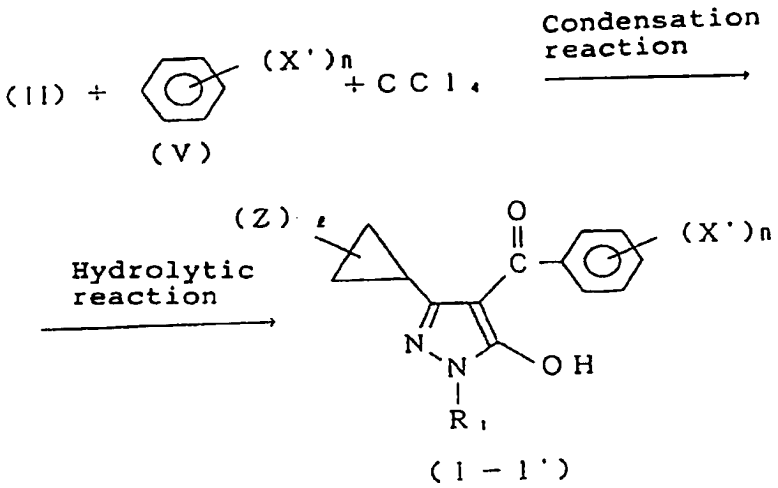
The pyrazole compounds of the formula (I) or their salts (hereinafter referred to as the compounds of the

present invention) can be prepared in accordance with the following reactions (A) to (E) and conventional methods for producing salts.

(A) When R_2 is a hydrogen atom:

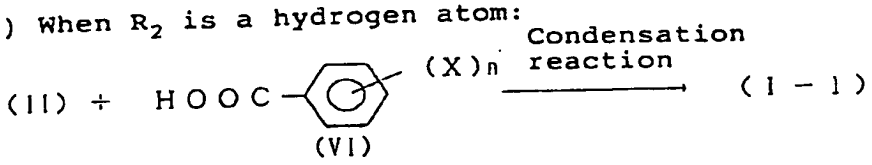


(B) When R_2 is a hydrogen atom:

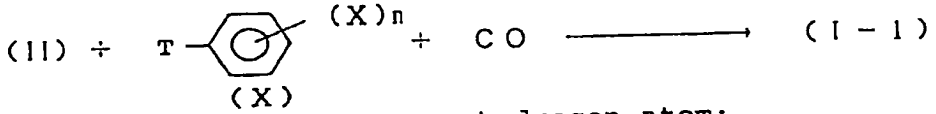


(I - 1')

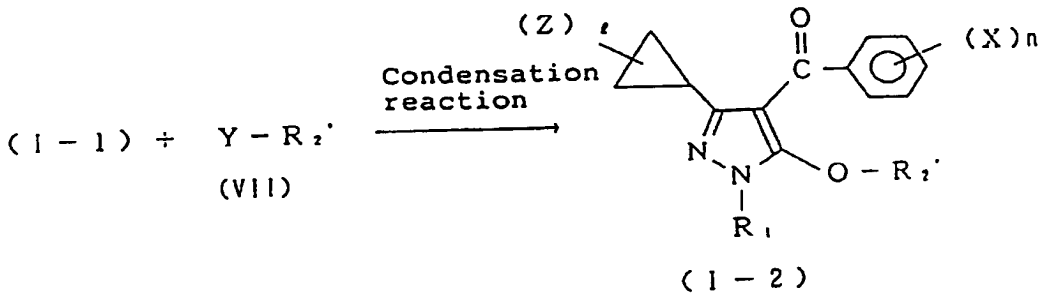
(C) When R_2 is a hydrogen atom:



(D) When R_2 is a hydrogen atom:



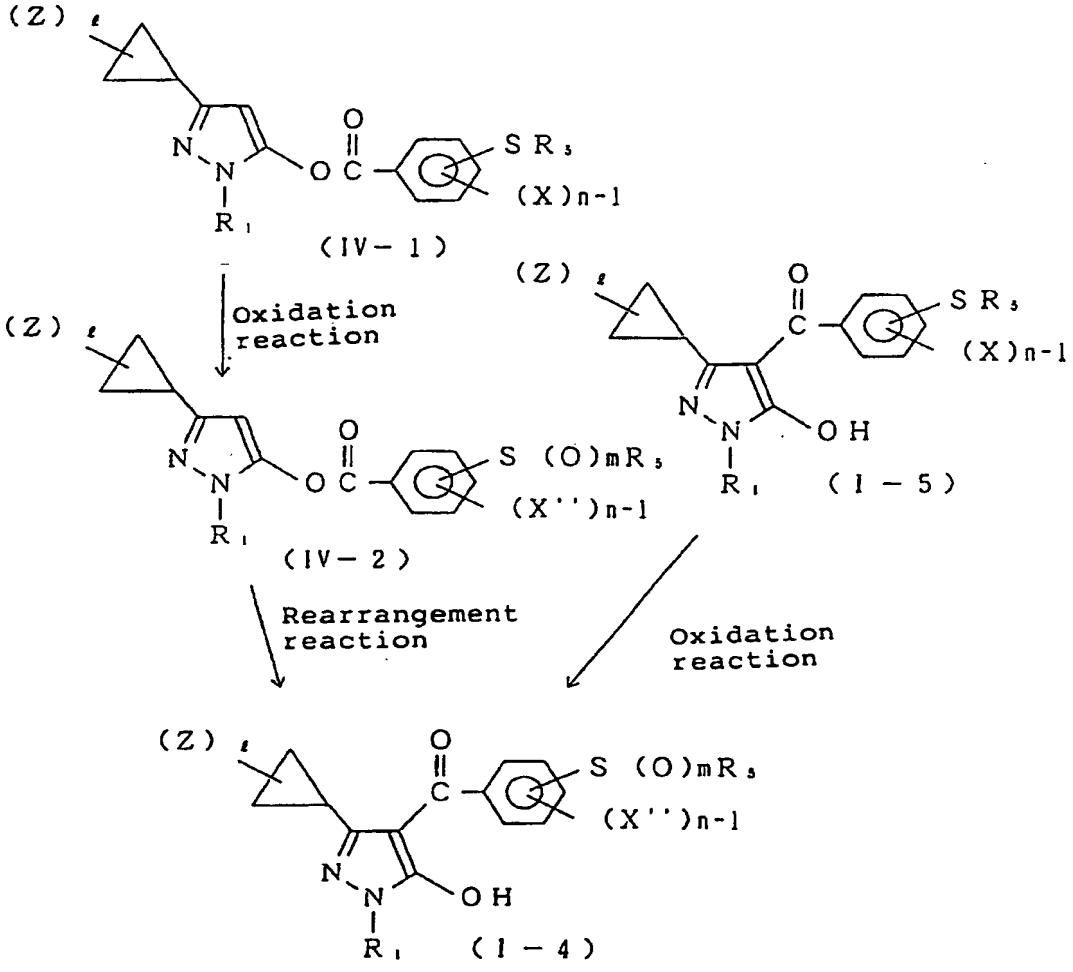
(E) When R_2 is other than a hydrogen atom:



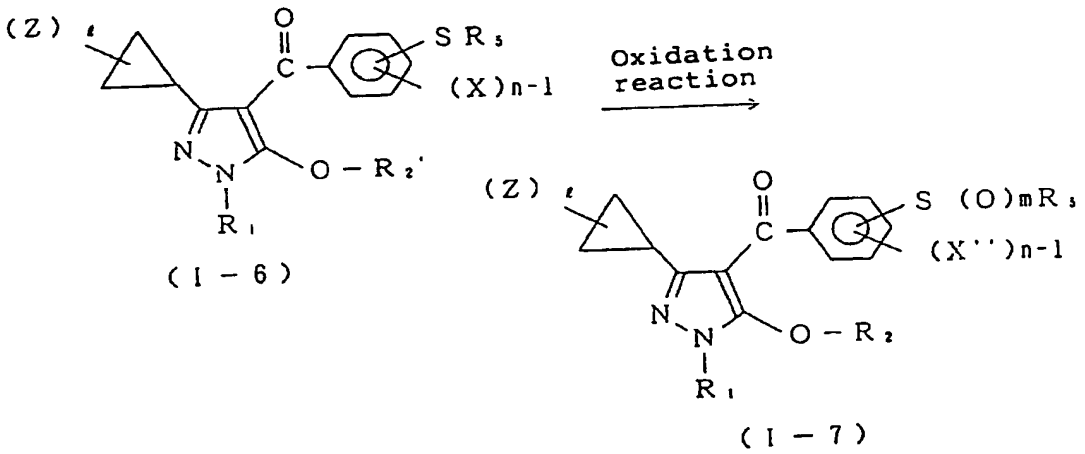
(I - 2)

Among the compounds of the present invention, those having certain predetermined substituents can be prepared in accordance with the following reactions (F) to (G) and conventional methods for preparing salts.

(F) When R_2 is a hydrogen atom, and (X)n contains at least one alkylsulfinyl or alkylsulfonyl group:



(G) When R_2 is other than a hydrogen atom, and (X)n contains at least one alkylsulfinyl or alkylsulfonyl group:



Now, the above reaction (A) will be described. In the reaction (A), R_1, X, Z, l and n are as defined above, and Y is a halogen atom.

The condensation reaction in the reaction (A) can be carried out, if necessary, in the presence of a base. As such a base, one or more members may be suitably selected for use from carbonates such as potassium carbonate and sodium carbonate; hydrogencarbonates such as potassium hydrogencarbonate and sodium hydrogencarbonate; metal hydrides such as potassium hydride and sodium hydride;

amines such as monomethylamine, dimethylamine and triethylamine; and pyridines such as pyridine and 4-dimethylaminopyridine.

Further, the condensation reaction in the reaction (A) can be carried out, if necessary, in the presence of a solvent. As such a solvent, any solvent may be used so long as it is a solvent inert to the reaction, and one or more members may be suitably selected for use from aromatic hydrocarbons such as benzene, toluene, xylene and chlorobenzene; cyclic or noncyclic aliphatic hydrocarbons such as carbon tetrachloride, methylene chloride, chloroform, dichloromethane, dichloroethane, trichloroethane, hexane and cyclohexane; ethers such as dioxane, tetrahydrofuran and diethyl ether; esters such as methyl acetate and ethyl acetate; polar aprotic solvents such as dimethylsulfoxide, sulfolane, dimethylacetamide, dimethylformamide, N-methylpyrrolidone, pyridine and hexamethylphosphoric triamide; nitriles such as acetonitrile, propionitrile and acrylonitrile; ketones such as acetone and methyl ethyl ketone; and water.

Further, the condensation reaction in the reaction (A) can be carried out, if necessary, in the presence of a phase transfer catalyst. As such a phase transfer catalyst, one or more members may be suitably selected for use from e.g. benzyltriethylammonium chloride, benzyltriethylammonium bromide, tetraethylammonium

chloride and tetraethylammonium bromide.

The reaction temperature of the condensation reaction in the reaction (A) is usually from 0 to 250°C, preferably from 15 to 150°C, and the reaction time is usually from 0.1 to 48 hours, preferably from 0.1 to 24 hours.

The compound of the formula (IV) which can be produced by the condensation reaction in this reaction (A), is a novel intermediate compound useful for producing the compounds of the present invention.

The rearrangement reaction in the reaction (A) comprises the following two steps i.e. (1) a rearrangement reaction step and (2) a pH adjusting reaction step. The rearrangement reaction step is carried out usually in the presence of a base. As such a base, one or more members may be suitably selected for use from carbonates such as potassium carbonate and sodium carbonate; and calcium hydroxide. The base is used usually in an amount of from 0.5 to 5 mols per mol of the compound of the formula (IV).

Further, the rearrangement reaction step of the rearrangement reaction in the reaction (A) can be carried out, if necessary, in the presence of a solvent. As such a solvent, any solvent may be used so long as it is a solvent inert to the reaction, and one or more members may suitably be selected for use from aromatic hydrocarbons such as benzene, toluene, xylene and

chlorobenzene; ethers such as dioxane, tetrahydrofuran and diethyl ether; and polar aprotic solvents such as dimethylsulfoxide, sulfolane, dimethylacetamide, dimethylforamide, N-methylpyrrolidone, pyridine and hexamethylphosphoric triamide.

The rearrangement reaction step of the rearrangement reaction in the reaction (A) is preferably carried out under an azeotropic dehydrating condition, whereby the rearrangement reaction will effectively proceed. This is one of preferred embodiments of the present invention. By the rearrangement reaction step, a salt of the compound of the formula (I) is produced, and a method for producing such a salt is also one of embodiments of the present invention. Further, a compound of the above-mentioned formula (I-2) can be produced by reacting a salt of the compound of the above formula (I) or a reaction mixture containing such a salt, obtained by this rearrangement reaction step, with a compound of the above formula (VII), under the reaction conditions for the reaction (D) which will be described hereinafter. This is also one of embodiments of the present invention.

The reaction temperature in the rearrangement reaction step is usually from 50 to 250°C, preferably from 50 to 150°C, and the reaction time is usually from 0.1 to 48 hours, preferably from 0.5 to 24 hours.

The pH adjusting reaction step of the rearrangement reaction in the reaction (A) is a reaction to adjust the

pH value to at most 7, which is carried out usually in the presence of an acidic substance and water. As such an acidic substance, one or more members may suitably be selected for use from inorganic acids such as hydrochloric acid and sulfuric acid; and organic acids such as acetic acid.

The pH adjusting reaction step of the rearrangement reaction in the reaction (A) can be carried out, if necessary, in the presence of a solvent. As such a solvent, any solvent may be used so long as it is a solvent inert to the reaction. For example, one or more members may be suitably selected for use from those mentioned in the description of the rearrangement reaction step as the preceding step.

The pH adjusting reaction step of the rearrangement reaction in the reaction (A) may be carried out after isolating the reaction product obtained by the rearrangement reaction step as the preceding step, in accordance with a conventional method, or may be carried out in one pot by using the reaction mixture obtained by the rearrangement reaction step, as it is. When it is carried out in one pot, it is carried out by adding and reacting an acidic substance and water to the reaction mixture obtained by the rearrangement reaction step as the preceding step.

The reaction temperature for the pH adjusting reaction step is usually from 0 to 100°C, preferably from

0 to 60°C.

Now, the above-mentioned reaction (B) will be described. In the reaction (B), R_1, Z, l, n and (II) are as defined above, and X' is a hydrogen atom, a halogen atom, an alkyl group, a haloalkyl group, an alkoxy group, an alkylthio group, an alkylsulfinyl group or an alkylsulfonyl group, provided that when n is at least 2, a plurality of X' may be the same or different.

The condensation reaction in the reaction (B) is carried out usually in the presence of a Lewis acid. As such a Lewis acid, one or more members may suitably be selected for use from e.g. dry aluminum chloride and dry aluminum bromide.

Further, the condensation reaction in the reaction (B) can be carried out, if necessary, in the presence of a solvent. As such a solvent, any solvent may be used so long as it is a solvent inert to the reaction, and one or more members may suitably be selected for use from halogenated aliphatic hydrocarbons such as carbon tetrachloride, methylene chloride, chloroform, dichloromethane, and dichloroethane.

The reaction temperature for the condensation reaction in the reaction (B) is usually from 0 to 80°C, and the reaction time is usually from 0.1 to 24 hours, preferably from 0.1 to 10 hours.

The hydrolytic reaction in the reaction (B) is carried out usually in the presence of an acidic

substance. As such an acidic substance, one or more members may suitably be selected for use from e.g. inorganic acids such as hydrochloric acid and sulfuric acid.

The hydrolytic reaction in the reaction (B) can be carried out, if necessary, in the presence of a solvent. As such a solvent, any solvent may be used so long as it is a solvent inert to the reaction, and one or more members may suitably be selected for use among those exemplified in the description of the condensation reaction as the preceding reaction.

The hydrolytic reaction in the reaction (B) may be carried out after isolating the reaction product obtained by the condensation reaction as the preceding reaction, in accordance with a conventional method, or may be carried out in one pot using the reaction mixture obtained by the condensation reaction as it is. In the case where it is carried out in one pot, post treatment such as removal of the Lewis acid may be applied, if necessary, to the reaction mixture obtained by the condensation reaction as the preceding reaction, and the acidic substance and water are added thereto to carry out the reaction.

The reaction temperature for the hydrolytic reaction in the reaction (B) is usually from 20 to 100°C, and the reaction time is usually from 0.1 to 24 hours, preferably from 0.1 to 10 hours.

Now, the above-mentioned reaction (C) will be described. In the reaction (C), X, n, (II) and (I-1) are as defined above.

The condensation reaction in the reaction (C) is carried out usually in the presence of a condensing agent and a solvent. As such a condensing agent, N,N'-dicyclohexylcarbodiimide may, for example, be mentioned, and as such a solvent, any solvent may be used so long as it is a solvent inert to the reaction, and one or more members may suitably be selected for use among alcohols such as tert-butyl alcohol and tert-amyl alcohol.

The condensation reaction in the reaction (C) can be carried out, if necessary, in the presence of a base. As such a base, one or more members may suitably be selected for use from e.g. carbonates such as potassium carbonate and sodium carbonate.

The reaction temperature for the condensation reaction in the reaction (C) is usually from 50 to 100°C, and the reaction time is usually from 0.1 to 24 hours, preferably from 0.5 to 20 hours.

Now, the above-mentioned reaction (D) will be described. In the reaction (D), X, n, (II) and (I-1) are as defined above, and T is a chlorine atom, a bromine atom or an iodine atom.

The reaction (D) is carried out usually in the presence of a base and a metal catalyst. As a base, one or more members may suitably be selected for use from

e.g. alkali metals such as sodium and potassium; alkali metal alkolates such as sodium methylate, sodium ethylate and potassium tert-butylate; carbonates such as potassium carbonate and sodium carbonate; hydrogencarbonates such as potassium hydrogencarbonate and sodium hydrogencarbonate; metal hydroxides such as potassium hydroxide and sodium hydroxide; metal hydrides such as potassium hydride and sodium hydride; amines such as monomethylamine, dimethylamine and triethylamine; pyridines such as pyridine and 4-dimethylaminopyridine; and N,N-dimethylaniline. As the metal catalyst, a transition metal such as palladium, rhodium, ruthenium or platinum, may be mentioned. The ligand used against the metal of the metal catalyst is not particularly limited, but an organophosphine compound such as triphenylphosphine or tri-n-butylphosphine is preferred.

The reaction (D) may be carried out, if necessary, in the presence of a solvent. As such a solvent, any solvent may be used so long as it is a solvent inert to the reaction. For example, one or more members may suitably be selected for use among aromatic hydrocarbons such as benzene, toluene, xylene and chlorobenzene; cyclic or noncyclic aliphatic hydrocarbons such as carbon tetrachloride, methylene chloride, chloroform, dichloromethane, dichloroethane, trichloroethane, hexane and cyclohexane; ethers such as dioxane, tetrahydrofuran and diethyl ether; esters such as methyl acetate and

ethyl acetate; polar aprotic solvents such as dimethylsulfoxide, sulfolane, dimethylacetamide, dimethylformamide, N-methylpyrrolidone and pyridine; nitriles such as acetonitrile, propionitrile and acrylonitrile; ketones such as acetone and methyl ethyl ketone; amines such as monomethylamine, dimethylamine and triethylamine; alcohols such as methanol, ethanol, propanol, and tert-butanol; organic acids such as acetic acid and propionic acid; aqueous ammonia; and water.

The reaction temperature for the reaction (D) is usually from 30 to 300°C, preferably from 50 to 200°C, and the reaction time is usually from 0.1 to 48 hours, preferably from 1 to 24 hours.

Now, the above-mentioned reaction (E) will be described. In the reaction (E), R_1, X, Y, Z, l, n and (I-1) are as defined above, and R_2' is a methyl group, $-A-R_3$, a phenyl group which may be substituted, a pyridyl group which may be substituted or an allyl group which is substituted by a phenyl group (wherein A and R_3 are as defined above).

The condensation reaction in the reaction (E) may be carried out, if necessary, in the presence of a base. As such a base, one or more members may suitably be selected for use from carbonates such as potassium carbonate and sodium carbonate; hydrogencarbonates such as potassium hydrogencarbonate and sodium hydrogencarbonate; metal hydroxides such as potassium hydroxide and sodium

hydroxide; metal hydrides such as potassium hydride and sodium hydride; amines such a monomethylamine, dimethylamine and triethylamine; and pyridines such as pyridine and 4-dimethylaminopyridine.

The condensation reaction in the reaction (E) may be carried out, if necessary, in the presence of a solvent. As such a solvent, any solvent may be used so long as it is inert to the reaction. For example, one or more members may suitably be selected for use from aromatic hydrocarbons such as benzene, toluene, xylene and chlorobenzene; cyclic or noncyclic aliphatic hydrocarbons such as carbon tetrachloride, methylene chloride, chloroform, dichloromethane, dichloroethane, trichloroethane, hexane and cyclohexane; ethers such as dioxane, tetrahydrofuran and diethyl ether; esters such as methyl acetate and ethyl acetate; polar aprotic solvents such as dimethylsulfoxide, sulfolane, dimethylacetamide, dimethylformamide, N-methylpyrrolidone, pyridine and hexamethylphosphoric triamide; nitriles such as acetonitrile, propionitrile and acrylonitrile; ketones such as acetone and methyl ethyl ketone; and water.

The condensation reaction in the reaction (E) may be carried out, if necessary, in the presence of a phase transfer catalyst and/or potassium iodide. As such a phase transfer catalyst, one or more members may suitably be selected for use among those mentioned for the

condensation reaction in the above-mentioned reaction (A).

The reaction temperature for the condensation reaction in the reaction (E) is usually from 0 to 200°C, preferably from 15 to 150°C, and the reaction time is usually from 0.1 to 48 hours, preferably from 0.1 to 24 hours.

Now, the above-mentioned reaction (F) will be described. In the reaction (F), R_1, X, Z, l and n are as defined above, R_5 is an alkyl group, preferably a C_{1-4} alkyl group, X" is a hydrogen atom, a halogen atom, an alkyl group, a haloalkyl group, an alkoxy group, an alkylsulfinyl group, an alkylsulfonyl group, a nitro group, an alkoxycarbonyl group, $-SO_2N(R_8)R_9$, $-N(R_{10})SO_2R_{11}$, $-CH_2S(O)_{q'}R_{12}$ or $-OSO_2R_{13}$, wherein R_8, R_9, R_{10}, R_{11}, R_{12} and R_{13} are as defined above, m is 1 or 2, and q' is 1 or 2. In the reaction (F), the oxidation reaction for producing (IV-2) from (IV-1) and the oxidation reaction for producing (I-4) from (I-5) (hereinafter referred to simply as the oxidation reaction) are carried out usually in the presence of an oxidizing agent and a solvent. As such an oxidizing agent, one or more members may suitably be selected for use from e.g. m-chloroperbenzoic acid and hydrogen peroxide. As the solvent, any solvent may be used so long as it is a solvent inert to the reaction. For example, one or more members may suitably be selected for

use among those mentioned for the condensation reaction in the above-mentioned reaction (B).

The reaction temperature for the oxidation reaction in the reaction (F) is usually from 0 to 80°C, and the reaction time is usually from 0.1 to 48 hours, preferably from 0.1 to 24 hours.

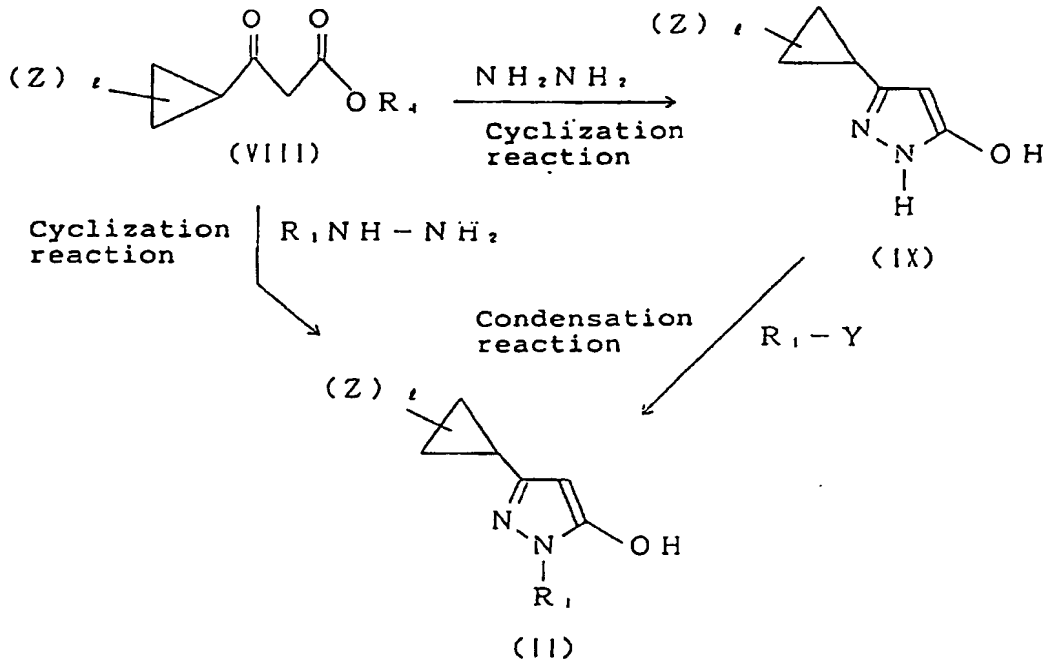
The rearrangement reaction in the reaction (F) can be carried out in accordance with the rearrangement reaction in the above-mentioned reaction (A).

Now, the above-mentioned reaction (G) will be described. In the reaction (G), R_1, R_2', R_5, X, X", Z, l, m and n are as defined above.

The oxidation reaction in the reaction (G) can be carried out in accordance with the oxidation reaction in the above-mentioned reaction (F).

The compound of the formula (II) in the above reactions (A), (B), (C) and (D) is a novel intermediate compound which is useful for producing the compounds of the present invention and may be produced, for example, by a method such as the reaction (H).

(H)



Now, the reaction (H) will be described. In the reaction (H), R_1, Y, Z and l are as defined above, and R_4 is a C_{1-6} alkyl group.

In the reaction (H), the cyclization reaction for producing (II) from (VIII) and the cyclization reaction for producing (IX) from (VIII) (hereinafter referred to simply as the cyclization reaction) may be carried out, if necessary, in the presence of a solvent. As such a solvent, any solvent may be used so long as it is a solvent inert to the reaction. For example, one or more members may suitably be selected for use from aromatic

hydrocarbons such as benzene, toluene, xylene and chlorobenzene; cyclic or noncyclic aliphatic hydrocarbons such as carbon tetrachloride, methylene chloride, chloroform, dichloromethane, dichloroethane, trichloroethane, hexane and cyclohexane; ethers such as dioxane, tetrahydrofuran and diethyl ether; polar aprotic solvents such as dimethylsulfoxide, sulfolane, dimethylacetamide, dimethylformamide, N-methylpyrrolidone pyridine and hexamethylphosphoric triamide; nitriles such as acetonitrile, propionitrile and acrylonitrile; and water.

The cyclization reaction in the reaction (H) may be carried out, if necessary, under an azeotropic dehydration condition.

The reaction temperature for the cyclization reaction in the reaction (H) is usually from 0 to 200°C, preferably from 20 to 150°C, and the reaction time is usually from 0.1 to 48 hours, preferably from 0.1 to 24 hours.

The condensation reaction in the reaction (H) is carried out usually in the presence of a base and a solvent. As the base, one or more members may suitably be selected for use from carbonates such as potassium carbonate and sodium carbonate; and metal hydrides such as potassium hydride and sodium hydride. Particularly preferred is potassium carbonate.

As the solvent, any solvent may be used so long as it

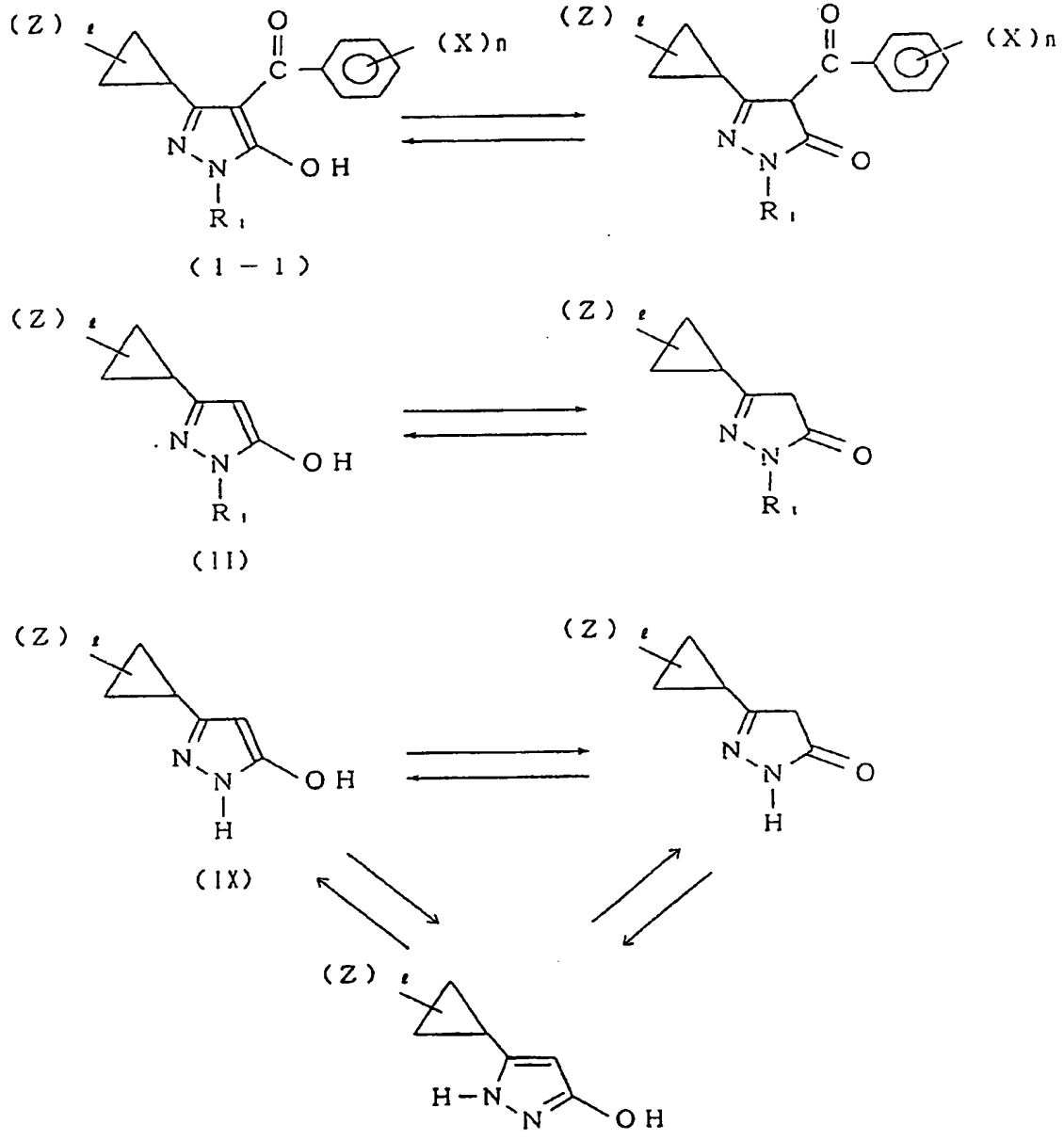
is a solvent inert to the reaction. For example, one or more members may suitably be selected for use from ethers such as dioxane, tetrahydrofuran and diethyl ether; and polar aprotic solvents such as dimethylsulfoxide, sulfolane, dimethylacetamide, dimethylformamide, N-methylpyrrolidone, pyridine and hexamethylphosphoric triamide. Particularly preferred is hexamethylphosphoric triamide.

The reaction temperature for the condensation reaction in the reaction (H) is usually from -20 to +150°C, preferably from -15 to +60°C, and the reaction time is usually from 0.1 to 24 hours, preferably from 0.1 to 10 hours.

The compound of the formula (IX) which can be prepared by the cyclization reaction in this reaction (H), is a novel intermediate compound which is useful for producing the compounds of the present invention.

The compounds of the present invention and the intermediate compounds useful for the production thereof, have the following isomers. Such various isomers (the respective isomers and mixtures of such isomers) are within the scope of the present invention.

(1) Among the compounds of the present invention represented by the above formula (I), compounds wherein R_2 is a hydrogen atom, and intermediate compounds represented by the above formulas (II) and (IX), have the following tautomers, respectively.



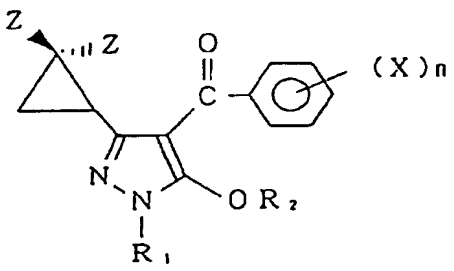
(I-1)

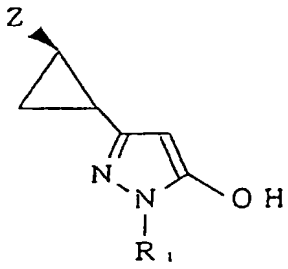
(II)

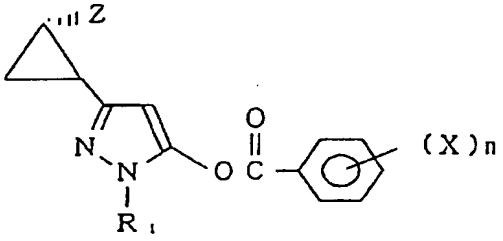
(IX)

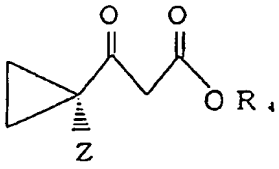
wherein R_1, X, Z, l and n are as defined above.

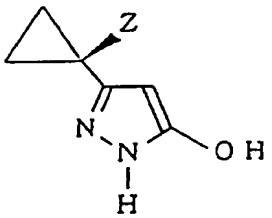
(2) Among the compounds of the present invention represented by the above formula (I) and the intermediate compounds represented by the above formulas (II), (IV), (VIII) and (IX), compounds wherein l is at least 1, have optical isomers. Some examples will be given below, but it should be understood that the optical isomers in the present invention are not limited to such specific examples.











wherein R_1, R_2, R_4, X, Z and n are as defined above.

In the specification of this application, such optical isomers are meant for a mixture of isomers (racemic modification) unless otherwise specified.

(3) Among compounds of the present invention represented by the above formula (I), compounds wherein R_2 is a $-A-R_3$, and R_3 is an alkenyl group which may be substituted, have geometrical isomers (E-isomer and Z-isomer).

The compound of the present invention exhibits excellent herbicidal effects when used as an active ingredient of a herbicide. It finds a wide range of applications to crop lands such as paddy fields, upland farms, orchards and mulberry fields, and non-crop lands such as forests, farm roads, playgrounds, and factory sites. The application method may suitably be selected from soil treatment application and foliar application.

The herbicidal composition containing the compound of the present invention is capable of controlling noxious weeds including grasses (or gramineae) such as barnyardgrass (Echinochloa crus-galli L.), crabgrass (Digitaria sanguinalis L.), greenfoxtail (Setaria viridis L.), goosegrass (Eleusine indica L.), wild oat (Avena fatua L.), johnsongrass (Sorghum halepense L.), quackgrass (Agropyron repens L.), alexandergrass (Brachiaria plantaginea), paragrass (Panicum purpurascens), sprangletop (Leptochloa chinensis) and red sprangletop (Leptochloa panicea); sedges (or Cyperaceae) such as rice flatsedge (Cyperus iria L.), purple nutsedge (Cyperus rotundus L.), japanese bulrush (Scirpus juncoides), flatsedge (Cyperus serotinus), small-flower

umbrellaplant (Cyperus difformis), slender spikerush (Eleocharis acicularis), and water chestnut (Eleocharis kuroguwai); alismataceae such as japanese ribbon wapato (Sagittaria pygmaea), arrow-head (Sagittaria trifolia) and narrowleaf waterplantain (Alisma canaliculatum); pontederiaceae such as monochoria (Monochoria vaginalis) and monochoria species (Monochoria korsakowii); scrophulariaceae such as false pimpernel (Lindernia pyxidaria) and abunome (Dopatrium junceum); lythraceae such as toothcup (Rotala indica) and red stem (Ammannia multiflora); and broadleaves such as velvetleaf (Abutilon theophrasti MEDIC.), tall morningglory (Ipomoea purpurea L.), common lambsquarters (Chenopodium album L.), prickly sida (Sida spinosa L.), common purslane (Portulaca oleracea L.), slender amaranth (Amaranthus viridis L.), redroot pigweed (Amaranthus retroflexus L.), sicklepod (Cassia obtusifolia L.), black nightshade (Solanum nigrum L.), pale smartweed (Polygonum lapathifolium L.), common cocklebur (Xanthium strumarium L.), flexuous bittercress (Cardamine flexuosa WITH.), henbit (Lamium amplexicaule L.) and threeseeded copperleaf (Acalypha australis L.).

Accordingly, it is useful for controlling noxious weeds non-selectively or selectively in the cultivation of a crop plant such as corn (Zea mays L.), soybean (Glycine max Merr.), cotton (Gossypium spp.), wheat (Triticum spp.), rice (Oryza sativa L.), barley (Hordeum

vulgare L.), oat (Avena sativa L.), sorgo (Sorghum bicolor Moench), rape (Brassica napus L.), sunflower (Helianthus annuus L.), sugar beet (Beta vulgaris L.), sugar cane (Saccharum officinarum L.), japanese lawngrass (Zoysia japonica stend), peanut (Arachis hypogaea L.) or flax (Linum usitatissimum L.) The compound of the present invention is particularly effective for selectively controlling noxious weeds in the cultivation of corn, wheat or rice, especially in the cultivation of corn.

The herbicidal composition containing the compound of the present invention is usually formulated by mixing the compound with various agricultural adjuvants and used in the form of a formulation such as a dust, granules, water-dispersible granules, a wettable powder, an emulsifiable concentrate, a water-based suspension concentrate, an oil-based suspension concentrate, water soluble granules (or powder), tablets or capsules. However, so long as it is suitable for the purpose of the present invention, it may be formulated into any type of formulation which is commonly used in this field.

Such agricultural adjuvants include solid carriers such as diatomaceous earth, slaked lime, calcium carbonate, talc, white carbon, kaoline, bentonite, a mixture of kaolinite and sericite, clay, sodium carbonate, sodium bicarbonate, mirabilite, zeolite and starch; solvents such as water, toluene, xylene, solvent

naphtha, dioxane, acetone, isophorone, methyl isobutyl ketone, chlorobenzene, cyclohexane, dimethylsulfoxide, dimethylformamide, N-methyl-2-pyrrolidone, and alcohol; anionic surfactants and spreaders such as a salt of fatty acid, a benzoate, an alkylsulfosuccinate, a dialkylsulfosuccinate, a polycarboxylate, a salt of alkylsulfuric acid ester, an alkyl sulfate, an alkylaryl sulfate, an alkyl diglycol ether sulfate, a salt of alcohol sulfuric acid ester, an alkyl sulfonate, an alkylaryl sulfonate, an aryl sulfonate, a lignin sulfonate, an alkyldiphenyl ether disulfonate, a polystyrene sulfonate, a salt of alkylphosphoric acid ester, an alkylaryl phosphate, a styrylaryl phosphate, a salt of polyoxyethylene alkyl ether sulfuric acid ester, a polyoxyethylene alkylaryl ether sulfate, a salt of polyoxyethylene alkyl ether sulfuric acid ester, a polyoxyethylene alkylaryl ether sulfuric acid ester, a polyoxyethylnee alkyl ether phosphate, a salt of polyoxyethylene alkyl aryl phosphoric acid ester, and a salt of a condensate of naphthalene sulfonate with formalin; nonionic surfactants and spreaders such as a sorbitan fatty acid ester, a glycerin fatty acid ester, a fatty acid polyglyceride, a fatty acid alcohol polyglycol ether, acetylene glycol, acetylene alcohol, an oxyalkylene block polymer, a polyoxyethylene alkyl ether, a polyoxyethylene alkylaryl ether, a polyoxyethylene styrylaryl ether, a polyoxyethylene glycol alkyl ether, a

polyoxyethylene fatty acid ester, a polyoxyethylene sorbitan fatty acid ester, a polyoxyethylene glycerin fatty acid ester, a polyoxyethylene hydrogenated castor oil, and a polyoxypropylene fatty acid ester; and vegetable and mineral oils such as olive oil, kapok oil, castor oil, palm oil, camellia oil, coconut oil, sesame oil, corn oil, rice bran oil, peanut oil, cottonseed oil, soybean oil, rapeseed oil, linseed oil, tung oil, and liquid paraffins. Such adjuvants may be selected for use among those known in this field, so long as the purpose of the present invention can thereby be accomplished. Further, various additives which are commonly used, such as a filler, a thickener, an anti-settling agent, an anti-freezing agent, a dispersion stabilizer, a phytotoxicity reducing agent, and an anti-mold agent, may also be employed.

The weight ratio of the compound of the present invention to the various agricultural adjuvants is usually from 0.1 :99.9 to 95:5, preferably from 0.2:99.8 to 85:15.

The dose of the herbicidal composition of the present invention can not generally be defined, since it may vary depending upon the weather condition, the soil condition, the type of the formulation, the types of the weeds to be controlled, the season for the application, etc. However, it is usually applied so that the compound of the present invention would be applied in an amount of

from 0.5 to 5000 g/ha, preferably from 1 to 1000 g/ha, more preferably from 5 to 500 g/ha. The present invention covers such a method for controlling noxious weeds by application of such a herbicidal composition.

The herbicidal compositions of the present invention may be used in admixture with or in combination with other agricultural chemicals, fertilizers or phytotoxicity-reducing agents. In such a case, they may exhibit even better effects or activities. As other agricultural chemicals, herbicides, fungicides, antibiotics, plant hormones or insecticides may, for example, be mentioned. Especially with a mixed herbicidal composition having the compound of the present invention used in admixture with or in combination with one or more active ingredients of other herbicides, it is possible to improve the herbicidal activities, the season for the application and the range of applicable weed types. Further, the compound of the present invention and an active ingredient of other herbicide may be separately formulated, so that they may be mixed for use at the time of application, or both may be formulated together. The present invention covers such mixed herbicidal compositions.

The blend ratio of the compounds of the present invention with the active ingredients of other herbicides can not generally be defined, since it varies depending upon the weather condition, the soil condition, the type

of the formulation, the season for the application, the manner of the application, etc. However, one active ingredient of other herbicide may be incorporated usually in an amount of from 0.001 to 10000 parts by weight, preferably from 0.01 to 1000 parts by weight, per part by weight of the compound of the present invention. Further, the total dose of all of the active ingredients is usually from 0.1 to 10000 g/ha, preferably from 0.2 to 5000 g/ha. The present invention covers a method for controlling noxious weeds by application of such herbicidal compositions.

As the active ingredients of other herbicides, the following (common names) may be mentioned.

(1) Those which are believed to exhibit herbicidal effects by disturbing auxin activities of plants, including a phenoxy acetic acid type such as 2,4-D, MCPA, MCPB or naproanilide, an aromatic carboxylic acid type such as 2,3,6-TBA, dicamba, picloram or clopyralid, and others such as benazolin, quinclorac, quinmerac or diflufenzopyr.

(2) Those which are believed to exhibit herbicidal effects by inhibiting photosynthesis of plants, including a urea type such as diuron, linuron, isoproturon or metobenzuron, triazine type such as simazine, atrazine, atratone, simetryn, prometryn, dimethametryn, metribuzin, terbuthylazine, cyanazine or ametryn, an uracil type such as bromacil or lenacil, an anilide type such as propanil

or cypromid, a carbamate type such as swep or phenmedipham, a hydroxybenzonitrile type such as bromoxynil, bromoxynil-octanoate or ioxynil, and others such as pyridate or bentazon.

(3) A quaternary ammonium salt type such as paraquat or diquat, which is believed to be converted to free radicals by itself to form active oxygen in the plant body and thus to exhibit quick herbicidal effects.

(4) Those which are believed to exhibit herbicidal effects by inhibiting chlorophyllbiosynthesis of plants and abnormally accumulating a photosensitizing peroxide substance in the plant body, including a diphenyl ether type such as nitrofen, chlomethoxyfen, bifenox, acifluorfen-sodium, fomesafen or oxyfluorfen, a cyclic imide type such as chlorphthalim, flumioxadine, flumiclorac-pentyl, methyl [2-chloro-4-fluoro-5-(5,6,7,8-tetrahydro-3-oxo-1H,3H-[1,3,4]thiadiazolo[3,4-a]pyridazin-1-ylideneamino)phenylthio] acetate (compound disclosed at page 60 of proceedings of 19th Meeting of Pesticide Science Society of Japan), and others such as oxadiation, sulfentrazone, carfentrazone-ethyl, thidiazimin, ethyl 2-chloro-5-(4-chloro-5-difluoromethoxyl-1-methylpyrazol-3-yl)-4-fluorophenoxyacetate (compound disclosed at pages 70-71 of proceedings of 21th Meeting of Pesticide Science Society of Japan).

(5) Those which are believed to exhibit herbicidal

effects characterized by whitening activities by inhibiting chromogenesis of plants such as carotenoids, including a pyridazinone type such as norflurazon or metflurazon, a pyrazole type such as pyrazolate, pyrazoxyfen or benzofenap, and others such as fluridone, flurtamone, diflufenican, methoxyphenone, clomazone, sulcotrione, 2-(2'-nitro-4'-methylsulfonyl-benzoyl)-1,3-cyclohexanedione (compound disclosed in US Patent 5,506,195), isoxaflutole or difenzoquat.

(6) Those which exhibit herbicidal effects specifically to gramineous plants, including an aryloxyphenoxypropionic acid type such as diclofop-methyl, pyriphenop-sodium, fluazifop-butyl, haloxyfop-methyl, quizalofop-ethyl or cyhalofop-butyl, and a cyclohexanedione type such as alloxydim-sodium, clethodim, sethoxydim or tralkoxydim.

(7) Those which are believed to exhibit herbicidal effects by inhibiting an amino acid biosynthesis of plants, including a sulfonylurea type such as chlorimuron-ethyl, sulfometuron-methyl, primisulfuron-methyl, bensulfuron-methyl, chlorsulfuron, metsulfuron-methyl, cinosulfuron, pyrazosulfuron-ethyl, azimsulfuron, flazasulfuron, rimusulfuron, nicosulfuron, imazosulfuron, cyclosulfamuron, prosulfuron, flupyrsulfuron, trisulfuron-methyl, halosulfuron-methyl or thifensulfuron-methyl, a triazolopyrimidinesulfoneamide type such as flumetsulam or metosulam, an imidazolinone

type such as imazapyr, imazethapyr, imazaquin, imazamox or imazameth or imazamethabenz, a pyrimidinylsalicylic acid type such as pyrithiobac-sodium, bispyribac-sodium or pyriminobac-methyl, and others such as glyphosate-ammonium, glyphosate-isopropylamine, glufosinate-ammonium or bialaphos.

(8) Those which are believed to exhibit herbicidal effects by inhibiting cell mitoses of plants, including a dinitroaniline type such as trifluralin, oryzalin, nitralin or pendimethalin, an organic phosphorus type such as amiprofos-methyl, butamifos, anilofos or piperophos, a phenylcarbamate type such as chlorpropham or barban, a cumylamine type such as daimuron, cumyluron or bromobutide, and others such as asulam or dithiopyr.

(9) Those which are believed to exhibit herbicidal effects by inhibiting protein biosynthesis or lipid biosynthesis of plants, including a thiocarbamate type such as EPTC, butylate, molinate, dimepiperate, esprocarb, thiobencarb or pyributicarb, or chloroacetamide type such as alachlor, butachlor, pretilachlor, metolachlor, thenylchlor, dimethenamid, acetochlor or propachlor, and other compounds such as a ethobenzanide, mefenacet, thiafluamide, tridiphane, cafenstrole, 4-(2-chlorophenyl)-N-cyclohexyl-4,5-dihydro-N-ethyl-5-oxo-1H-tetrazol-1-carboxyamide (compound disclosed in JP-A-6-306061), oxaziclomefon, or 2-ethyl-2-[2-(3-chlorophenyl)-2,3-epoxypropyl]-indan-1,3-dione

(compound disclosed in JP-A-2-304043).

As is evident from Test Examples 1 and 2 given hereinafter, the compound of the present invention include those which show selectivity for effectively controlling weeds, while showing safety to crop plants such as rice, wheat and corn. When the compound of the present invention is to be used in the cultivation of such crop plants, synergistic effects may be obtained by using it in admixture with or in combination with one or more of the following compounds among the above-mentioned active compounds of other herbicides.

In the cultivation of rice:

2,4-D, MCPA, MCPB, naproanilide, quinclorac, simetryn, prometryn, dimethametryn, propanil, swep, bentazon, nitrofene, chlomethoxyfen, bifenox, oxadiazon, pyrazolate, pyrazoxyfen, benzofenap, methoxyphenone, cyhalofop-butyl, bensulfuron-methyl, cinosulfuron, pyrazosulfuron-ethyl, azimsulfuron, imazosulfuron, cyclosulfamuron, bispyribac-sodium salt, pyriminobac-methyl, anilofos, piperophos, daimuron, cumyluron, bromobutide, dithiopyr, molinate, dimepiperate, esprocarb, thiobencarb, pyributicarb, thenylchlor, pretilachlor, butachlor, ethobenzanide, mefenacet, cafenstrole, 4-(2-chlorophenyl)-N-cyclohexyl-4,5-dihydro-N-ethyl-5-oxo-1H-tetrazole-1-carboxyamide, oxaziclomefon, and 2-ethyl-2-[2-(3-chlorophenyl)-2,3-epoxypropyl]-indane-1,3-dione.

In the cultivation of corn:

2,4-D, MCPA, dicamba, clopyralid, benazolin, diflufenzopyr, diuron, linuron, metobenzuron, simazine, atrazine, atratone, metribuzin, terbuthylazine, cyanazine, ametryn, cypromid, bromoxynil, bromoxynil-octanoate, pyridate, bentazon, paraquat, oxyfluorfen, flumiclorac-pentyl, methyl [2-chloro-4-fluoro-5-(5,6,7,8-tetrahydro-3-oxo-1H,3H-[1,3,4]thiadiazolo[3,4-a]pyridazin-1-ylideneamino)phenylthio] acetate, fluridone, sulcotrione, 2-(2'-nitro-4'-methylsulfonylbenzoyl)-1,3-cyclohexanedione, isoxaflutole, carfentrazone ethyl, primisulfuron methyl, rimusulfuron, nicosulfuron, prosulfuron, halosulfuron-methyl, thifensulfuron-methyl, flumetsulam, metosulam, imazethapyr, glyphosate-ammonium salt, glyphosate-isopropyl amine salt, glufosinate-ammonium salt, trifluralin, pendimethalin, EPTC, butylate, alachlor, metolachlor, acetochlor, propachlor, dimethenamid and tridiphane.

In the cultivation on wheat:

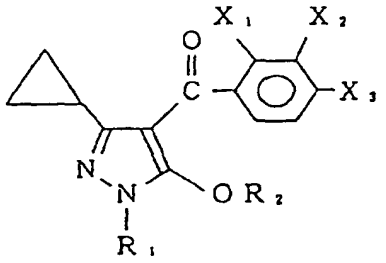
MCPB, quinmerac, linuron, isoproturon, prometryn, bromoxynil, bromoxynil-octanoate, pyridate, bifenox, carfentrazone-ethyl, thidiazimin, ethyl 2-chloro-5-(4-chloro-5-difluoromethoxyl-1-methylpyrazol-3-yl)-4-fluorophenoxy acetate, flurtamone, diflufenican, sulcotrione, diclofop-methyl, tralkoxydim, chlorsulfuron, metsulfuron-methyl, prosulfuron, halosulfuron-methyl,

flumetsulam, metosulam, pendimethalin, barban and imazamethabenz.

Now, preferred embodiments of the present invention will be described.

(1) The pyrazole compound of the above formula (I) or its salt.

(2) The pyrazole compound or its salt according to Item 1, wherein the formula (I) is represented by the formula (I'):



(I')

wherein R_1 is an alkyl group, R_2 is a hydrogen atom or $-A-R_3$, A is $-SO_2-$, $-CO-$, $-CH_2-$ or $-CH_2CO-$, R_3 is an alkyl group which may be substituted, an alkenyl group which may be substituted, an alkynyl group which may be substituted , a cyano group or a phenyl group which may be substituted, each of X^1, X^2 and X^3 is a hydrogen atom, a halogen atom, an alkyl group, a haloalkyl group, an alkoxy group, an alkylthio group, an alkylsulfinyl group, an alkylsulfonyl group, a nitro group, $-SO_2N(R_8)R_9$, $-N(R_{10})SO_2R_{11}$, $-CH_2S(O)qR_{12}$ or $-OSO_2R_{13}$, each of R_8, R_9, R_{10}, R_{11}, R_{12} and R_{13} is an alkyl group, and q is an integer of from 0 to 2.

(3) The pyrazole compound or its salt according to Item 2, wherein A is $-SO_2-$, $-CH_2-$ or $-CH_2CO-$, each of X^1, X^2 and X^3 is a hydrogen atom, a halogen atom, an alkyl group, a haloalkyl group, an alkoxy group, an alkylthio group, an alkylsulfinyl group, an alkylsulfonyl group or a nitro group.

(4) The pyrazole compound or its salt according to Item 3, wherein X^1 is an alkylthio group, an alkylsulfinyl group or an alkylsulfonyl group, and each of X^2 and X^3 is a hydrogen atom, a halogen atom, an alkyl group, a haloalkyl group or a nitro group.

(5) A herbicide containing the pyrazole compound or its salt as defined in Item 1, 2, 3, or 4, as an active ingredient.

(6) A method for controlling noxious weeds, which comprises applying an effective amount of the pyrazole compound or its salt as defined in Item 1, 2, 3, or 4.

(7) A method for controlling noxious weeds, which comprises applying an effective amount of the pyrazole compound or its salt as defined in Item 1, 2, 3, or 4 to an upland field.

(8) A method for controlling noxious weeds, which comprises applying an effective amount of the pyrazole compound or its salt as defined in Item 1, 2, 3, or 4 to a corn field.

(9) A method for controlling noxious weeds, which comprises applying an effective amount of the pyrazole

compound or its salt as defined in Item 1, 2, 3, or 4 to a wheat field.

(10) A method for controlling noxious weeds, which comprises applying an effective amount of the pyrazole compound or its salt as defined in Item 1, 2, 3, or 4 to a paddy field.

(11) A mixed herbicidal composition comprising at least one member selected from the pyrazole compound or its salt as defined in Item 1, 2, 3, or 4 and at least one member selected from active ingredient compounds of other herbicides.

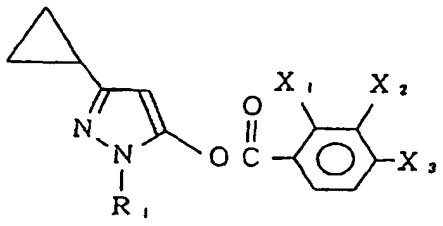
(12) The compound of the above formula (II).

(13) The compound according to Item 12, wherein l is 0.

(14) The compound of the above formula (IV).

(15) The compound according to Item 14, wherein l is 0.

(16) The compound according to Item 14, wherein the formula (IV) is represented by the formula (IV'):



(IV')

wherein R_1 is an alkyl group, each of X^1, X^2 and X^3 is a hydrogen atom, a halogen atom, an alkyl group, a haloalkyl group, an alkoxy group, an alkylthio group, an alkylsulfinyl group, an alkylsulfonyl group, a nitro group, $-SO_2N(R_8)R_9$, $-N(R_{10})SO_2R_{11}$, $-CH_2S(O)qR_{12}$ or $-OSO_2R_{13}$, each of R_8, R_9, R_{10}, R_{11}, R_{12} and R_{13} is an alkyl group, and q is an integer of from 0 to 2.

(17) The pyrazole compound or its salt according to Item 16, wherein each of X^1, X^2 and X^3 is a hydrogen atom, a halogen atom, an alkyl group, a haloalkyl group, an alkoxy group, an alkylthio group, an alkylsulfinyl group, an alkylsulfonyl group or a nitro group.

(18) The pyrazole compound or its salt according to Item 17, wherein X^1 is an alkylthio group, an alkylsulfinyl group or an alkylsulfonyl group, and each of X^2 and X^3 is a hydrogen atom, a halogen atom, an alkyl group, a haloalkyl group or a nitro group.

BEST MODE FOR CARRYING OF THE INVENTION

Now, the present invention will be described in further detail with reference to Examples. However, it should be understood that the present invention is by no means restricted to such specific Examples. Firstly, Preparation Examples for the compounds of the present invention will be described.

PREPARATION EXAMPLE 1

Preparation of 3-cyclopropyl-4-(4-trifluoromethyl-2-methylsulfonylbenzoyl)-5-hydroxy-1-methylpyrazole (after-

mentioned Compound No. a-11) and 3-cyclopropyl-4-(4-trifluoromethyl-2-methylsulfonylbenzoyl)-1-methyl-5-pyrazolyl p-toluene sulfonate (after-mentioned Compound No. a-12) (First method)

1) 1.4 g of methylhydrazine was added at room temperature to a solution having 5.53 g of tert-butyl 3-cylocopropyl-3-oxopropionate dissolved in 30 ml of tetrahydrofuran, and the mixture was reacted for about 2 hours under reflux.

After completion of the reaction, tetrahydrofuran was distilled off under reduced pressure to obtain 4.14 g of crude 3-cyclopropyl-5-hydroxy-1-methylpyrazole (after-mentioned Intermediate No. 1a-1).

The melting point of this product was from 95 to 121°C, and the NMR spectrum data are as follows.

^{1}H-NMR δppm [Solvent: $CDCl_3$]

0.76-0.8 (m,2H), 0.9-0.99 (m,2H), 1.74-1.81 (m,1H), 3.06 (s), 3,26 (s,3H), 4.6 (bs)

2) A solution having 0.41 g of sodium carbonate dissolved in 30 ml of water, was added to a solution having 1 g of 3-cyclopropyl-5-hydroxy-1-methylpyrazole obtained in the preceding step dissolved in 30 ml of toluene, followed by stirring for 5 minutes. Then, 4-trifluoromethyl-2-methylthiobenzoyl chloride preliminarily prepared by mixing and reacting under reflux for one hour, 1.52 g of 4-trifluoromethyl-2-methylthiobenzoic acid, 5 ml of thionyl chloride and a

catalytic amount of N,N-dimethylformamide, followed by removal of excess thionyl chloride, was added thereto, and the mixture was reacted at 50°C for one hour.

After completion of the reaction, the reaction mixture was cooled and put into water, and extracted with ethyl acetate. The obtained ethyl acetate layer was washed with a saturated sodium chloride aqueous solution and then dried over anhydrous sodium sulfate, and the solvent was distilled off under reduced pressure. The obtained residue was purified by silica gel column chromatography to obtain 0.8 g of oily 3-cyclopropyl-1-methyl-5-pyrazolyl 4-trifluoromethyl-2-methylthiobenzoate (after-mentioned Intermediate No. 2a-16). The NMR spectrum data of the product are as follows.

^{1}H-NMR δppm [Solvent: $CDCl_3$]

0.69-0.73 (m,2H), 0.86-0.91 (m,2H), 1.85-1.92 (m,1H), 2.53 (s,3H), 3.70 (s,3H), 5.94 (s,1H), 7.46 (d,1H), 7.53 (s,1H), 8.24 (d,1H)

3) 0.91 g of methachloroperbenzoic acid was dividedly added at room temperature to a solution having 0.75 g of 3-cyclopropyl-1-methyl-5-pyrazolyl 4-trifluoromethyl-2-methylthiobenzoate obtained in the preceding step dissolved in 30 ml of methylene chloride, and the mixture was reacted for one hour within a range of from room temperature to 40°C.

After completion of the reaction, the reaction mixture was put into water and extracted with methylene

chloride.

The obtained methylene chloride layer was washed with dilute alkali and then with water, and thereafter dried over anhydrous sodium sulfate, and methylene chloride was distilled off. The obtained residue was purified by silica gel column chromatography to obtain 0.75 g of 3-cyclopropyl-1-methyl-5-pyrazolyl 4-trifluoromethyl-2-methylsulfonylbenzoate (after-mentioned Intermediate No. 2a-5) having a melting point of from 99 to 102°C. The NMR spectrum data of the product are as follows.

^{1}H-NMR δppm [Solvent: $CDCl_3$]

0.73-0.77 (m,2H), 0.86-0.94 (m,2H), 1.87-1.93 (m,1H), 2.05 (s,3H), 3,74 (s,3H), 5.95 (s,1H), 8.0 (d,1H), 8.06 (d,1H), 8.47 (s,1H)

4) A mixture comprising 0.7 g of 3-cyclopropyl-1-methyl-5-pyrazolyl 4-trifluoromethyl-2-methylsulfonylbenzoate obtained in the preceding step, 0.3 g of dry potassium carbonate, 25 ml of toluene and 5 ml of N,N-dimethylformamide, was reacted for one hour under an azeotropic dehydration condition using a Dean-Stark azeotropic dehydration apparatus.

After completion of the reaction, the reaction mixture was cooled and put into water, and the aqueous layer was washed with ethyl acetate. The aqueous layer was acidified with concentrated hydrochloric acid and extracted with ethyl acetate. The obtained ethyl acetate layer was washed with a saturated sodium chloride aqueous

solution and dried over anhydrous sodium sulfate. Then, ethyl acetate was distilled off under reduced pressure to obtain the desired product 3-cyclopropyl-4-(4-trifluoromethyl-2-methylsulfonylbenzoyl)-5-hydroxy-1-methylpyrazole (after-mentioned Compound No. a-11) as a viscous crude product. The NMR spectrum data of this product are as follows.

^{1}H-NMR δppm [Solvent: $CDCl_3$]

0.42-0.45 (m,2H), 0.72-0.81 (m,2H), 0.95-1.05 (m,1H), 3.34 (s,3H), 3.67 (s,3H), 7.73 (d,1H), 8.0 (d,1H), 8.4 (s,1H)

The melting point of 3-cyclopropyl-4-(4-trifluoromethyl-2-methylsulfonylbenzoyl)-5-hydroxy-1-methylpyrazole as the above-mentioned viscous crude product, was from 83 to 93°C.

5) 0.155 g of p-toluene sulfonyl chloride was added to a mixture comprising 0.3 g of 3-cyloropropyl-4-(4-trifluoromethyl-2-methylsulfonylbenzoyl)-5-hydroxy-1-methylpyrazole obtained in the preceding step, 0.118 g of dry potassium carbonate, 0.002 g of tetraethyl ammonium bromide, 20 ml of toluene and 5 ml of N,N-dimethylformamide, and the mixture was reacted for about one hour within a range of from 40 to 50°C with stirring.

After completion of the reaction, the reaction mixture was put into water and extracted with ethyl acetate. The obtained ethyl acetate layer was washed with water and further with a saturated sodium chloride

aqueous solution and then dried over anhydrous sodium sulfate, and ethyl acetate was distilled off under reduced pressure. The obtained residue was purified by silica gel column chromatography to obtain 0.3 g of 3-cyclopropyl-4-(4-trifluoromethyl-2-methylsulfonylbenzoyl)-1-methyl-5-pyrazolyl p-toluene sulfonate (after-mentioned Compound No. a-12) as a viscous desired product. The NMR spectrum data of this product are as follows.

^{1}H-NMR δppm [Solvent: $CDCl_3$]

0.52-0.56 (m,2H), 0.8-0.84 (m,2H), 1.48-1.55 (m,1H), 2.47 (s,3H), 3.32 (s,3H), 3.65 (s,3H), 7.37 (d,2H), 7.58 (d,1H), 7.82 (d,2H), 7.89 (d,1H), 8.28 (s,1H)

The melting point of the above viscous 3-cyclopropyl-4-(4-trifluoromethyl-2-methylsulfonylbenzoyl)-1-methyl-5-pyrazolyl p-toluene sulfonate was from 67 to 70°C.

PREPARATION EXAMPLE 2

Preparation of 3-cyclopropyl-4-(4-trifluoromethyl-2-methylsulfonylbenzoyl)-5-hydroxy-1-methylpyrazole (after-mentioned Compound No. a-11) and 3-cyclopropyl-4-(4-triflouromethyl-2-methylsulfonylbenzoyl)-1-methyl-5-pyrazolyl p-toluene sulfonate (after-mentioned compound No. a-12) (Second method)

1) A mixture comprising 3.88 g of 3-cyloropropyl-1-methyl-5-pyrazolyl 4-trifluoromethyl-2-methylsulfonylbenzoate (after-mentioned Intermediate No. 2a-5), 1.52 g of dry potassium carbonate, 100 ml of

toluene and 20 ml of N,N-dimethylformamide, was reacted for one hour under an azeotropic dehydration condition using a Dean-Stark azeotropic dehydration apparatus.

After completion of the reaction, the reaction mixture was cooled and put into water, followed by liquid separation. The obtained aqueous layer was acidified with concentrated hydrochloric acid and extracted with ethyl acetate. The obtained ethyl acetate layer was washed with water and then with a saturated sodium chloride aqueous solution and dried over anhydrous sodium sulfate. Then, ethyl acetate was distilled off under reduced pressure to obtain 3.88 g of viscous 3-cyclopropyl-4-(4-trifluoromethyl-2-methylsulfonylbenzoyl)-5-hydroxy-1-methylpyrazole as a crude product. This product was left to stand to sufficiently remove the solvent to obtain crystals of 3-cyclopropyl-4-(4-trifluoromethyl-2-methylsulfonylbenzoyl)-5-hydroxy-1-methylpyrazole (after-mentioned Compound No. a-11) as the desired product, having a melting point of from 153 to 157°C.

2) 0.36 g of p-toluene sulfonyl chloride was added to a mixture comprising 0.7 g of crystals of 3-cyclopropyl-4-(4-trifluoromethyl-2-methylsulfonylbenzoyl)-5-hydroxy-1-methylpyrazole obtained in the preceding step, 0.27 g of dry potassium carbonate, 0.005 g of tetraethylammonium bromide, 20 ml of toluene and 4 ml of N,N-dimethylformamide, and the mixture was reacted for about

1.5 hours within a range of from 40 to 50°C with stirring.

After completion of the reaction, the reaction mixture was put into water and extracted with ethyl acetate. The obtained ethyl acetate layer was washed with a saturated sodium chloride aqueous solution and dried over anhydrous sodium sulfate. Then, ethyl acetate was distilled off under reduced pressure. The obtained residue was purified by silica gel column chromatography to obtain 0.7 g of crystals of 3-cyclopropyl-4-(4-triflouromethyl-2-methylsulfonylbenzoyl)-1-methyl-5-pyrazolyl p-toluene sulfonate (after-mentioned Compound No. a-12) as the desired product, having a melting point of from 135 to 138°C.

PREPARATION EXAMPLE 3

Preparation of 3-cyclopropyl-4-(2,4-dichloro-3-methylbenzoyl)-5-hydroxy-1-methylpyrazole (after-mentioned Compound No. a-8)

1) 2.76 g of 3-cyclopropyl-5-hydroxy-1-methylpyrazole (after-mentioned Intermediate No. 1a-1) and 3.9 g of 2,6-dichlorotoluene were charged into 30 ml of 1,2-dichloroethane, and 6.7 g of dry aluminum chloride was dividedly added thereto at a temperature of not higher than 50°C with stirring. After the addition, stirring was continued for from 10 to 15 minutes within a range of from 35 to 40°C. Then, a solution having 4.0 g of carbon tetrachloride dissolved in 4 ml of 1,2-dichloroethane,

was dropwise added thereto at the same temperature. After completion of the dropwise addition, the mixture was reacted for 1.5 hours at a temperature of from 40 to 45°C.

After completion of the reaction, the reaction mixture was put into 150 ml of ice water to separate a 1,2-diclhoroethane layer.

2) 0.5 ml of water was added thereto, and the mixture was heated to 50°C, whereupon 3.5 ml of concentrated sulfuric acid was gradually dropwise added thereto. After completion of the dropwise addition, the mixture was reacted for 1.5 hours under reflux.

After completion of the reaction, the reaction mixture was left to cool, and 150 ml of water was added thereto, followed liquid separation. The obtained 1,2-dichloroethane layer was washed with water and then extracted with an alkaline solution having 3.5 g of sodium hydroxide dissolved in 100 ml of water. Then, 50% sulfuric acid was added thereto to make the liquid weakly acidic and extracted with methylene chloride. The obtained methylene chloride layer was dried over anhydrous sodium sulfate, and methylene chloride was distilled off under reduced pressure to obtain 3.5 g of the desired product having a melting point of from 112 to 115°C. The NMR spectrum data of this product are as follows.

^{1}H-NMR δppm [Solvent: $CDCl_3$]

0.66-0.71 (m,2H), 0.93-0.99 (m,2H), 1.15-1.22 (m,1H), 2.72 (s,3H), 3.89 (s,3H), 7.34 (d,1H), 7.57 (d,1H)

Preparation Example 4

Preparation of 4-(2,4-dichlorobenzoyl)-3-cyclopropyl-1-ethyl-5-hydroxypyrazole (after-mentioned Compound No. a-18)

1) A solution having 0.87 g of dry hydrazine dissolved in 5 ml of dry tetrahydrofuran, was added to a solution having 5 g of tert-butyl 3-cyclopropyl-3-oxopropionate dissolved in 30 ml of dry tetrahydrofuran, and the mixture was reacted for one hour under reflux.

After completion of the reaction, tetrahydrofuran, etc. were distilled off under reduced pressure to obtain 3.3 g of 3-cyclopropyl-5-hydroxypyrazole (after-mentioned Intermediate No. 3-1) having a melting point of from 213 to 217°C. The NMR spectrum data of this product are as follows.

^{1}H-NMR δppm [Solvent: heavy MDSO]

0.57-0.61 (m,2H), 0.81-0.86 (m,2H), 1.70-1.77 (m,1H), 5.1 (s,1H), 10.16 (bs,1H)

2) 1.61 g of 3-cyclopropyl-5-hydroxypyrazole obtained in the preceding step was mixed with a solution having 1.89 g of dry potassium carbonate dissolved in 20 ml of hexamethylphosphoric triamide, and the mixture was cooled within a range of from 0 to 2°C. Then, iodoethane was dropwise added thereto within a range of from 0 to 5°C over a period of about 15 minutes. Then, the mixture was

reacted for one hour at the same temperature and then further reacted for one hour within a range of from room temperature to 40°C.

3) 2.72 g of 2,4-dichlorobenzoyl chloride was added thereto at room temperature, and the mixture was reacted for 0.5 hour at the same temperature and further reacted for 0.5 hour at 40°C.

After completion of the reaction, the reaction mixture was put into water and extracted with toluene. The obtained toluene layer was thoroughly washed with water and then with a saturated sodium chloride aqueous solution and then dried over anhydrous sodium sulfate. Then, toluene was distilled off under reduced pressure, and the obtained residue was purified by silica gel column chromatography to obtain 1.2 g of 3-cyclopropyl-1-ethyl-5-pyrazolyl 2,4-dichlorobenzoate (after-mentioned Intermediate No. 2a-7) having a melting point of from 61 to 63°C. The NMR spectrum data of this product are as follows.

^{1}H-NMR δppm [Solvent: $CDCl_3$]

0.69-0.73 (m,2H), 0.87-0.9 (m,2H), 1.4 (t,3H), 1.85-1.92 (m,1H), 4.02-4.08 (q,2H), 5.92 (s,1H), 7.39 (d,1H), 7.55 (s,1H), 7.94 (d,1H)

4) Using 1.1 g of 3-cyclopropyl-1-ethyl-5-pyrazolyl 2,4-dichlorobenzoate obtained in the preceding step, 0.843 g of the desired product having a melting point of from 74 to 77°C was obtained in the same manner as Step

4) in Preparation Example 1.

PREPARATION EXAMPLE 5

Preparation of 3-cyclopropyl-4-(4-trifluoromethyl-2-methylsulfonylbenzoyl)-1-methyl-5-pyrazolyl benzene sulfonate (after-mentioned Compound No. a-27)

1) 0.57 g of 3-cyclopropyl-1-methyl-5-pyrazolyl 4-trifluoromethyl-2-methylsulfonyl benzoate (after-mentioned Intermediate No. 2a-5), 20 ml of toluene and 1 ml of N,N-dimethylformamide were charged into an Erlenmeyer flask, and 0.11 g of potassium carbonate was added thereto. The mixture was reacted for 15 hours under an azeotropic dehydration condition to obtain a reaction mixture containing a potassium salt of 3-cyclopropyl-4-(4-trifluoromethyl-2-methylsulfonylbenzoyl)-5-hydroxy-1-methylpyrazole.

2) The reaction mixture obtained in the preceding step was left to cool, and 0.1 g of tetraethylammonium chloride and 0.1 g of potassium iodide were added thereto. Then, 0.27 g of benzene sulfonyl chloride was added thereto. The mixture was reacted for 5.5 hours at 55°C with stirring.

After completion of the reaction, the reaction mixture was put into water and extracted with ethyl acetate. Then, the extract was washed with water. The obtained organic layer was dried over anhydrous sodium sulfate, then concentrated and thereafter purified by silica gel column chromatography to obtain 0.49 g of the

desired product having a melting point of from 175 to 178°C. The NMR spectrum data of this product are as follows.

^{1}H-NMR δppm [Solvent: $CDCl_3$]

0.46-0.05 (m,2H), 0.73-0.81 (m,2H), 1.33-1.41 (m,1H), 3.27 (s,3H), 3.63 (s,3H), 7.53-7.58 (m,3H), 7.7 (t,1H), 7.85 (d,1H), 7.96 (d,2H), 8.27 (s,1H)

PREPARATION EXAMPLE 6

Preparation of 3-cyclopropyl-4-(4-trifluoromethyl-2-methylsulfonylbenzoyl)-1-methyl-5-pyrazolyl n-propanesulfonate (after-mentioned Compound No. a-89)

A mixture comprising 0.4 g of 3-cyclopropyl-4-(4-trifluoromethyl-2-methylsulfonylbenzoyl)-5-hydroxy-1-methylpyrazole (after-mentioned Compound No. a-11), 20 ml of toluene, 5 ml of N,N-dimethylformamide, 5 mg of tetraethylammonium bromide and 0.16 g of n-propanesulfonyl chloride, was reacted for about 12 hours at 40°C with stirring.

After completion of the reaction, the reaction mixture was put into water and extracted with ethyl acetate. The obtained ethyl acetate layer was washed with a saturated sodium chloride aqueous solution and then dried over anhydrous sodium sulfate. Then, ethyl acetate was distilled off under reduced pressure. The obtained residue was purified by silica gel column chromatography to obtain 0.34 g of the desired product having a melting point of from 128 to 131°C. The NMR

spectrum data of this product are as follows.

^{1}H-NMR δppm [Solvent: $CDCl_3$]

0.43-0.51 (m,2H), 0.78-0.82 (m,2H), 1.12 (t,3H), 1.1-1.2 (m,1H), 2.0-2.1 (m,2H), 3.33 (s,3H), 3.53 (t,2H), 3.82 (s,3H), 7.70 (d,1H), 7.96 (d,1H), 8.38 (s,1H)

PREPARATION EXAMPLE 7

Preparation of 5-benzyloxy-3-cyclopropyl-4-(4-trifluoromethyl-2-methylsulfonylbenzoyl)-1-methylpyrazole (after-mentioned Compound No. a-94)

0.14 g of benzyl chloride was added to a mixture comprising 0.4 g of 3-cyclopropyl-4-(4-trifluoromethyl-2-methylsulfonylbenzoyl)-5-hydroxy-1-methylpyrazole (after-mentioned Compound No. a-11), 0.16 g of dry potassium carbonate, 5 mg of benzyltriethylammonium chloride, 5 mg of potassium iodide, 20 ml of toluene and 5 ml of N,N-dimethylformamide, and the mixture was reacted for 24 hours within a range of from 50 to 70°C with stirring.

After completion of the reaction, the reaction mixture was put into water and extracted with ethyl acetate. The obtained ethyl acetate layer was washed with a saturated sodium chloride aqueous solution and then dried over anhydrous sodium sulfate. Then, ethyl acetate was distilled off under reduced pressure. The obtained residue was purified by silica gel column chromatography to obtain 0.25 g of the desired product having a melting point of from 154 to 157°C. The NMR spectrum data of this product are as follows.

^{1}H-NMR δppm [Solvent: $CDCl_3$]

0.68-0.71 (m,2H), 0.85-0.88 (m,2H), 1.8-2.0 (m,1H), 3.35 (s,3H), 3.42 (s,3H), 5.00 (s,2H), 7.11-7.12 (m,2H), 7.26-7.30 (m,3H), 7.58-7.60 (d,1H), 7.58-7.88 (d,1H), 8.34 (s,1H)

PREPARATION EXAMPLE 8

Preparation of 5-(2-chloro-2-propenyloxy)-3-cyclopropyl-4-(4-trifluoromethyl-2-methylsulfonylbenzoyl)-1-methylpyrazole (after-mentioned Compound No. a-213)

A mixture comprising 0.776 g of 3-cyclopropyl-4-(4-trifluoromethyl-2-methylsulfonylbenzoyl)-5-hydroxy-1-methylpyrazole (after-mentioned Compound No. a-11), 30 ml of toluene, 4 ml of N,N-dimethylformamide, 5 mg of tetraethylammonium bromide and 0.245 g of 2,3-dichloropropene, was reacted for 1 hour at room temperature, then reacted for 4 hours at a temperature of from 60 to 80°C with stirring.

After completion of the reaction, the reaction mixture was put into water and extracted with ethyl acetate. The obtained ethyl acetate layer was washed with a saturated sodium chloride aqueous solution and then dried over anhydrous sodium sulfate. Then, ethyl acetate was distilled off. The obtained residue was purified by silica gel column chromatography to obtain 0.65 g of the desired product having a melting point of from 180 to 111°C. The NMR spectrum data of this product are as follows.

^{1}H-NMR δppm [Solvent: $CDCl_3$]

0.59-0.61 (m,2H), 0.84-0.86 (m,2H), 1.6-1.7 (m,1H), 3.38 (s,3H), 3.67 (s,3H), 4.68 (s,2H), 5.37-5.4 (d,2H), 7.62 (d,1H), 7.93 (d,1H), 8.38 (s,1H)

PREPARATION EXAMPLE 9

Preparation of 3-cyclopropyl-1-methyl-4-(2-methylthio-4-trifluoronmethylbenzoyl)-5-hydroxypyrazole (after-mentioned Compound No. a-82), 3-cyclopropyl-1-methyl-4-(2-methylthio-4-trifluoromethylbenzoyl)-5-pyrazolyl p-toluene sulfonate (after-mentioned Compound No. a-72) and 3-cyclopropyl-4-(4-trifluoromethyl-2-methylsulfonylbenzoyl)1-methyl-5-pyrazolyl p-toluene sulfonate (after-mentioned Compound No. a-12)

1) Into a 200 ml autoclave, 1.59 g of 4-iodo-3-methylthiobenzotrifluoride prepared in accordance with the following Preparation Example 10, 1.38 g of 3-cyclopropyl-5-hydroxy-1-methylpyrazole (after-mentioned Intermediate No. 1a-1), 0.5 g of triethylamine, 3.1 g of potassium carbonate, 0.22 g of palladium (II) bis(triphenylphosphine) dichloride and 40 ml of dioxane were put and sealed, and the interior of the autoclave was flushed with carbon monoxide (pressure: 65 kg/cm^2), followed by a reaction at 140°C for 8 hours. After completion of the reaction, the solvent was distilled off, and the residue was dissolved in water, and then insoluble matters were filtered off. The filtrate was washed with dichloromethane. The washed product was

acidified (pH=1) with concentrated hydrochloric acid and extracted with dichloromethane. The obtained extract solution was dried over anhydrous sodium sulfate, and the solvent was distilled off to obtain 1.59 g of 3-cyclopropyl-1-methyl-4-(2-methylthio-4-trifluoromethylbenzoyl)-5-hydroxypyrazole (after-mentioned Compound No. a-82) as a reddish brown solid.

2) 1.59 g of 3-cyclopropyl-1-methyl-4-(2-methylthio-4-trifluoromethylbenzoyl)-5-hydroxypyrazole obtained in the preceding step was mixed, without purification, with 20 ml of toluene, 4 ml of N,N-dimethylformamide, 0.94 g of p-toluene sulfonyl chloride and 0.34 g of potassium carbonate, and the mixture was reacted at 60°C for 3 hours. After completion of the reaction, water was added to the reaction mixture, and the mixture was extracted with ethyl acetate. The extract solution was dried over anhydrous sodium sulfate, and the solvent was distilled off. The obtained residue was purified by silica gel column chromatography (developing solvent: ethyl acetate/hexane=1/4) to obtain 0.53 g of 3-cyclopropyl-1-methyl-4-(2-methylthio-4-trifluoromethylbenzoyl)-5-pyrazolyl p-toluene sulfonate (after-mentioned Compound No. a-72). The NMR spectrum data of this product are as follows.

^{1}H-NMR δppm [Solvent: $CDCl_3$]

0.79 (m,2H), 0.90 (m,2H), 1.97 (m,1H), 2.39 (s,3H), 2.47 (s,3H), 7.23 (d,2H), 7.32 (d,1H), 7.48 (s,1H), 7.49

(d,1H), 7.53 (d,2H)

3). 0.46 g of 3-cyclopropyl-1-methyl-4-(2-methylthio-4-trifluoromethylbenzoyl)-5-pyrazolyl p-toluene sulfonate obtained in the preceding step was dissolved in 10 ml of dichloromethane, and 0.47 g of 85% methachloroperbenzoic acid was added thereto under cooling with ice. Then, the mixture was returned to room temperature and reacted over night with stirring. After completion of the reaction, an aqueous sodium hydrogen carbonate solution was added to the reaction mixture, and the mixture was extracted with dichloromethane. The extract layer was dried over anhydrous sodium sulfate, and the solvent was distilled off. The obtained residue was purified by silica gel column chromatography (developing solvent: ethyl acetate/hexane=3/7) to obtain 0.49 g of 3-cyclopropyl-4-(4-trifluoromethyl-2-methylsulfonylbenzoyl)-1-methyl-5-pyrazolyl p-toluene sulfonate (after-mentioned Compound No. a-12).

PREPARATION EXAMPLE 10

Preparation of 4-iodo-3-methylthiobenzotrifluoride

1) 123.85 g of sodium iodide was added to a solution having 42.23 g of 4-chloro-3-nitrobenzotrifluoride dissolved in 200 ml of N,N-dimethylformamide, and the mixture was reacted at 140°C for 17 hours.

After completion of the reaction, the reaction mixture was put into water and extracted with ethyl ether. The ethyl ether layer was washed with water and

then dried over anhydrous sodium sulfate. Then, ethyl ether was distilled off. The obtained residue was purified by silica gel column chromatography to obtain 44.15 g of 4-iodo-3-nitrobenzotrifluoride. The NMR spectrum data of this product are as follows.

^{1}H-NMR δppm [Solvent: $CDCl_3$]

7.52 (dd,1H), 8.11 (s,1H), 8.22 (d,1H)

2) A solution having 30 g of 4-iodo-3-nitrobenzotrifluoride obtained in the preceding step dissolved in 300 ml of acetic acid, was heated, and 26.43 g of reduced iron was added thereto over a period of 15 minutes at a temperature of from 85 to 95°C. Then, the mixture was reacted for further 5 minutes at the same temperature.

After completion of the reaction, the reaction mixture was cooled with ice, and insoluble matters were filtered off using celite. The filtration cake was thoroughly washed with ethyl acetate, and the washing liquid and the filtrate were mixed, followed by washing with water for 5 times. The obtained ethyl acetate layer was dried over anhydrous sodium sulfate, and ethyl acetate was distilled off under reduced pressure. The obtained residue was purified by silica gel column chromatography to obtain 25.52 g of oily 3-amino-4-iodobenzotrifluoride. The NMR spectrum data of this product are as follows.

^{1}H-NMR δppm [Solvent: $CDCl_3$]

6.70 (dd,1H), 6.93 (d,1H), 7.73 (d,1H)

3) To a solution containing a part (5.1 g) of 3-amino-4-iodebenzotrifluoride obtained in the preceding step, 16.75 g of dimethyldisulfide and 80 ml of chloroform, a solution having the rest (20.42 g) of 3-amino-4-iodebenzotrifluoride obtained in the preceding step dissolved in 20 ml of chloroform and 11.92 g of tert-butylnitrite, were simultaneously dropwise added at a temperature of from 25 to 30°C. After completion of the dropwise addition, the mixture was reacted at room temperature for 16 hours.

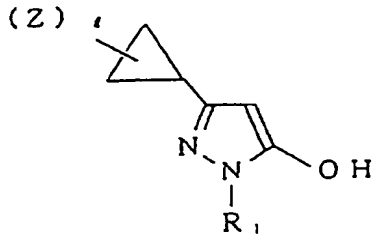
After completion of the reaction, 200 ml of methylene chloride was added to the reaction mixture, and the mixture was washed with an aqueous hydrochloric acid solution with pH 1 to 2. Then, the methylene chloride layer was washed with water and dried over anhydrous sodium sulfate. Then, methylene chloride and chloroform were distilled off under reduced pressure. The obtained residue was purified by silica gel column chromatography to obtain 19.89 g of the desired product as an oily substance. The NMR spectrum data of this product are as follows.

^{1}H-NMR δppm [Solvent: $CDCl_3$]

2.51 (s,3H), 7.08 (dd,1H), 7.26 (d,1H), 7.90 (dd,1H)

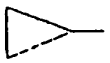
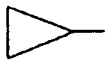
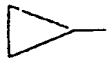
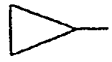
Other compounds of the present invention can be prepared in accordance with the above described Preparation Examples or the above described various

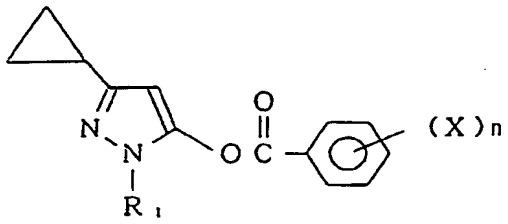
processes for producing the compounds of the present invention. Typical examples of the intermediate compound represented by the above formula (II) will be shown in Table 1, typical examples of the intermediate compound represented by the above formula (IV) will be presented in Tables 2a and 2b, typical examples of the intermediate compound represented by the above formula (IX) will be presented in Table 3, and typical examples of the compound of the present invention represented by the above formula (I) will be presented in Tables 4a and 4b.



... (II)

Table 1

Intermediate No.	R_1	(Z) 	Physical properties
1a-1	CH_3		m.p. 95-121°C
1a-2	CH_2CH_3		
1a-3	n-C_3H_7		
1a-4	n-C_4H_9		
1a-5	$CH(CH_3)_2$		
1b-1	CH_3		
1b-2	CH_2CH_3		
1b-3	n-C_3H_7		
1b-4	n-C_4H_9		
1b-5	$CH(CH_3)_2$		



… (IVa)

Table 2a

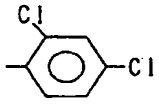
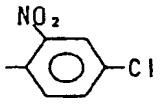
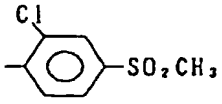
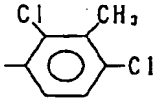
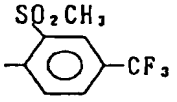
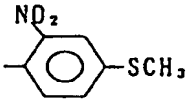
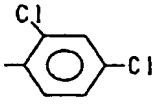
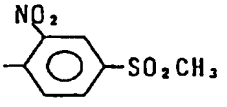
Intermediate No.	R_1	(X)n	Physical properties
2a-1	CH_3		
2a-2	CH_3		m.p. 84—87℃
2a-3	CH_3		m.p. 148—150℃
2a-4	CH_3		
2a-5	CH_3		m.p. 99—102℃
2a-6	CH_3		
2a-7	CH_2CH_3		m.p. 61—63℃
2a-8	CH_3		

Table 2a (continued)

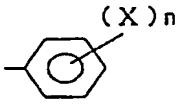
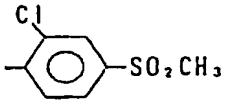
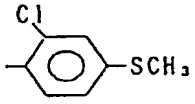
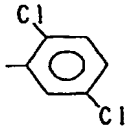
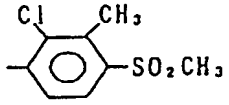
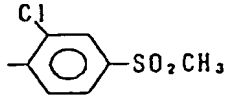
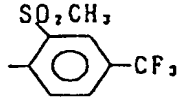
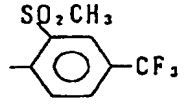
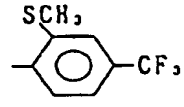
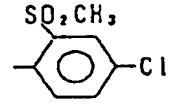
Intermediate No.	R_1		Physical properties
2a-9	CH_2CH_3		m.p. 96—99°C
2a-10	CH_3		Viscous
2a-11	CH_3		
2a-12	CH_2CH_3		
2a-13	$n\text{-}C_3H_7$		
2a-14	CH_2CH_3		m.p. 94—97°C
2a-15	$CH(CH_3)_2$		
2a-16	CH_3		Oily
2a-17	CH_3		m.p. 136—140 °C

Table 2a (continued)

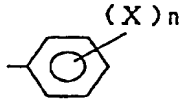
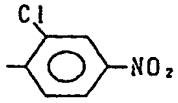
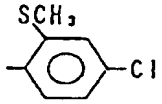
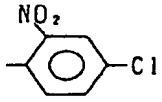
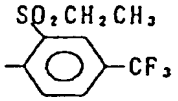
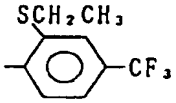
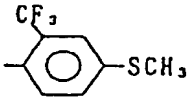
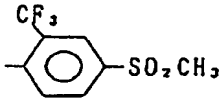
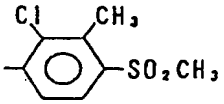
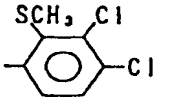
Inter-mediate	R_1		Physical properties
2 a − 18	CH_3		
2 a − 19	CH_3		m.p. 90−93℃
2 a − 20	CH_2CH_3		
2 a − 21	CH_3		Viscous
2 a − 22	CH_3		Viscous
2 a − 23	CH_3		Viscous
2 a − 24	CH_3		m.p. 146 −149 ℃
2 a − 25	CH_3		m.p. 125 − 130 ℃
2 a − 26	CH_3		

Table 2a (continued)

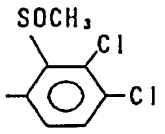
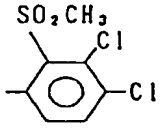
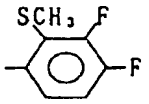
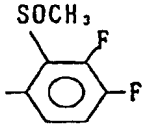
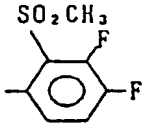
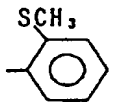
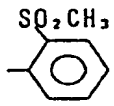
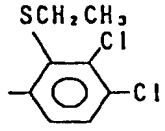
Intermediate No.	R_1	(X)n	Physical properties
2a-27	CH_3		
2a-28	CH_3		m.p. 134 −136 ℃
2a-29	CH_3		
2a-30	CH_3		
2a-31	CH_3		
2a-32	CH_3		
2a-33	CH_3		
2a-34	CH_3		

Table 2a (continued)

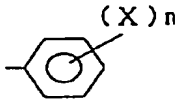
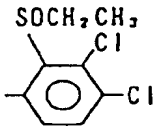
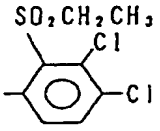
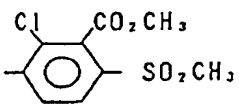
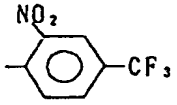
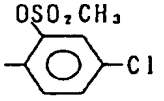
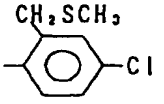
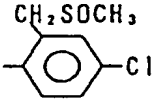
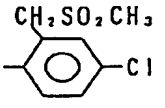
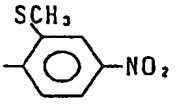
Intermediate No.	R_1		Physical properties
2 a − 35	CH_3		
2 a − 36	CH_3		
2 a − 37	CH_3		
2 a − 38	CH_3		
2 a − 39	CH_3		
2 a − 40	CH_3		
2 a − 41	CH_3		
2 a − 42	CH_3		
2 a − 43	CH_3		m.p. 118 − 122 ℃

Table 2a (continued)

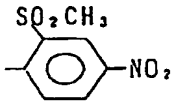
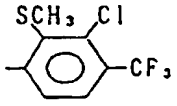
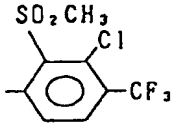
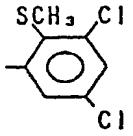
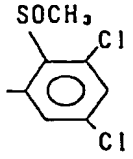
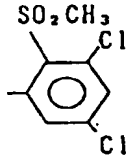
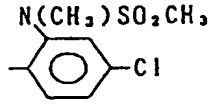
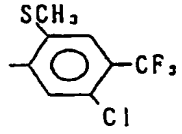
Intermediate No.	R_1	(X)n	Physical properties
2a-44	CH_3		
2a-45	CH_3		Viscous
2a-46	CH_3		
2a-47	CH_3		
2a-48	CH_3		
2a-49	CH_3		
2a-50	CH_3		
2a-51	CH_3		

Table 2a (continued)

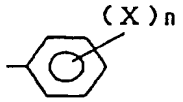
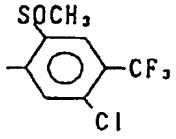
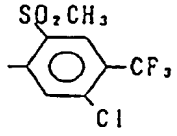
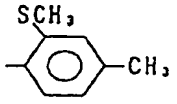
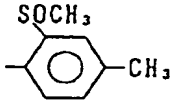
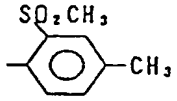
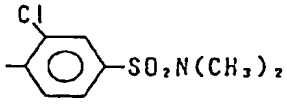
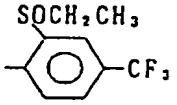
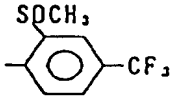
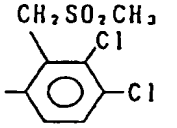
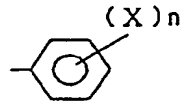
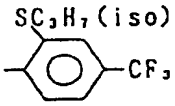
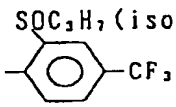
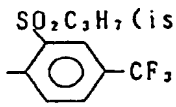
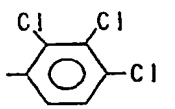
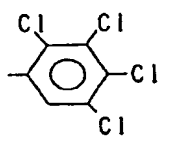
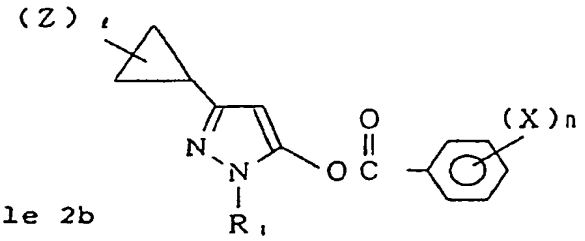
Intermediate No.	R_1		Physical properties
2a-52	CH_3		
2a-53	CH_3		
2a-54	CH_3		
2a-55	CH_3		
2a-56	CH_3		
2a-57	CH_3		
2a-58	CH_3		
2a-59	CH_3		
2a-60	CH_3		

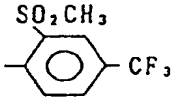
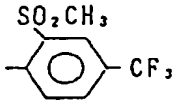
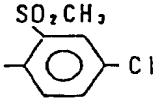
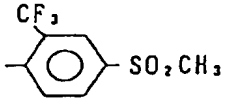
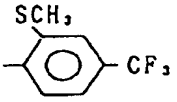
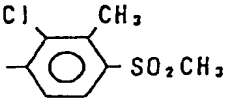
Table 2a (continued)

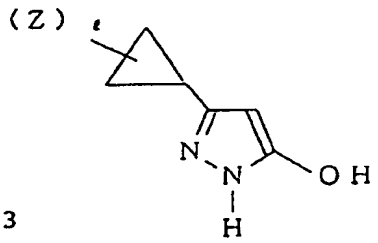
Intermediate No.	R_1		Physical properties
2a-61	CH_3		
2a-62	CH_3		
2a-63	CH_3		
2a-64	CH_3		
2a-65	CH_3		



... (IV)

Table 2b

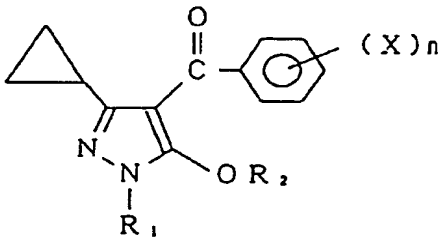
Intermediate No.	R_1	(Z) t	(X)n	Physical properties
2 b − 1	CH_3			Viscous
2 b − 2	CH_2CH_3			
2 b − 3	CH_3			
2 b − 4	CH_3			
2 b − 5	CH_3			
2 b − 6	CH_3			



… (IX)

Table 3

Intermediate No.	(Z) t	Physical properties
3 – 1		m.p. 213 – 217 ℃
3 – 2		



... (I a)

Table 4a

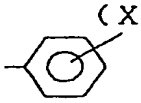
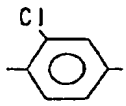
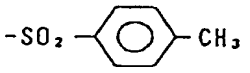
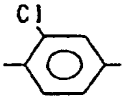
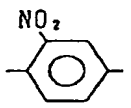
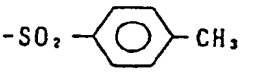
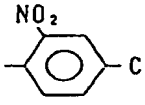
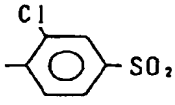
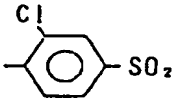
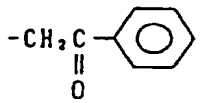
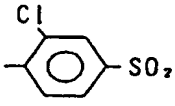
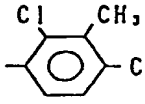
Compound No.	R_1	R_2		Physical properties
a − 1	CH_3	H		m.p. 131− 133℃
a − 2	CH_3			Refractive index $n_D^{43.4}$ 1.5779
a − 3	CH_3	H		m.p. 65—70℃
a − 4	CH_3			m.p. 130—133℃
a − 5	CH_3	H		m.p. 163− 166℃
a − 6	CH_3			m.p. 172− 174℃
a − 7	CH_3			m.p. 145− 147℃
a − 8	CH_3	H		m.p. 112− 115℃

Table 4a (continued)

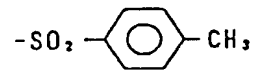
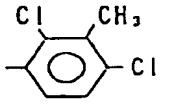
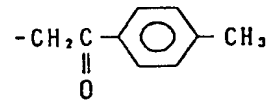
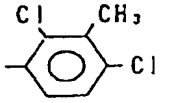
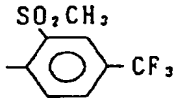
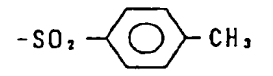
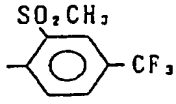
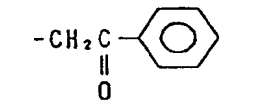
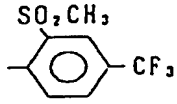
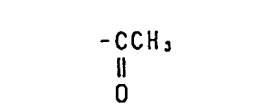
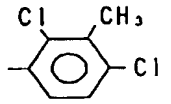
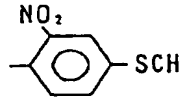
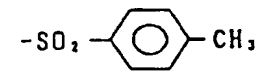
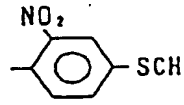
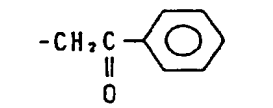
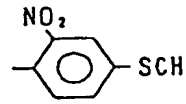
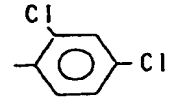
Compound No.	R_1	R_2	(X)n	Physical properties
a − 9	CH_3			m.p. 115 - 118℃
a − 10	CH_3			m.p. 126 - 129℃
a − 11	CH_3	H		m.p. 153 - 157℃
a − 12	CH_3			m.p. 135 - 138℃
a − 13	CH_3			m.p. 124 - 127℃
a − 14	CH_3			m.p. 112 - 115℃
a − 15	CH_3	H		m.p. 115 - 122℃
a − 16	CH_3			m.p. 146 - 148℃
a − 17	CH_3			Viscous
a − 18	CH_2CH_3	H		m.p. 74 - 77℃

Table 4a (continued)

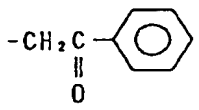
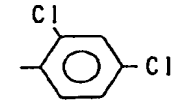
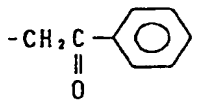
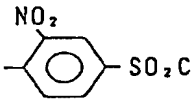
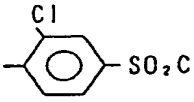
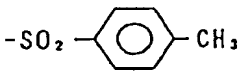
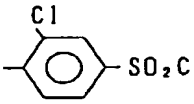
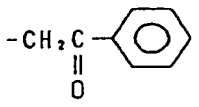
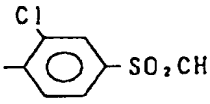
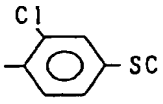
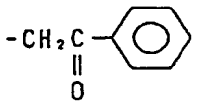
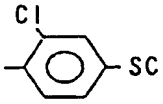
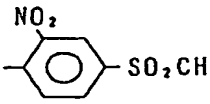
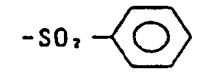
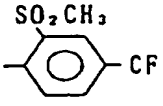
Compound No.	R_1	R_2	(X)n	Physical properties
a-19	CH_2CH_3			Viscous
a-20	CH_3			m.p. 181-183°C
a-21	CH_2CH_3	H		m.p. 158-161°C
a-22	CH_2CH_3			m.p. 116-118°C
a-23	CH_2CH_3			m.p. 146-148°C
a-24	CH_3	H		m.p. 111-114°C
a-25	CH_3			Refractive index $n_D^{16.6}$ 1.6001
a-26	CH_3	H		m.p. 140-145°C
a-27	CH_3			m.p. 175-178°C

Table 4a (continued)

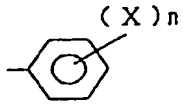
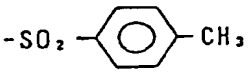
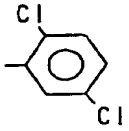
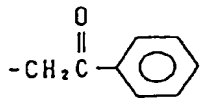
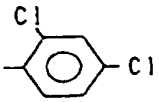
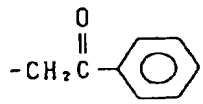
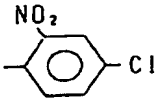
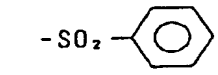
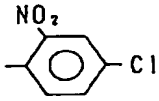
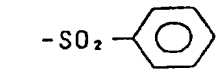
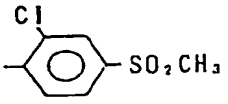
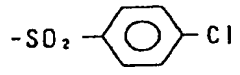
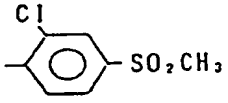
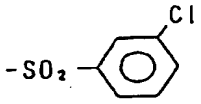
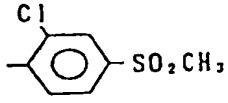
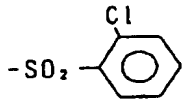
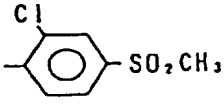
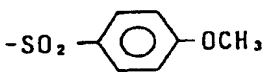
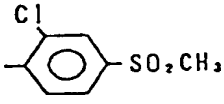
Compound No.	R_1	R_2		Physical properties
a − 28	CH_3			
a − 29	CH_3			
a − 30	CH_3			
a − 31	CH_2CH_3			
a − 32	CH_2CH_3			
a − 33	CH_3			
a − 34	CH_3			
a − 35	CH_3			
a − 36	CH_3			

Table 4a (continued)

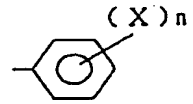
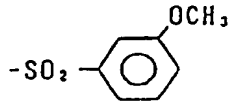
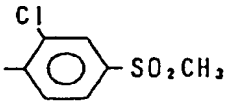
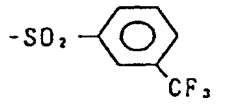
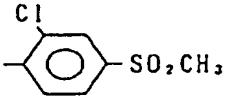
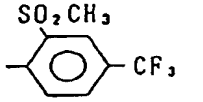
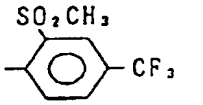
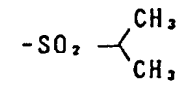
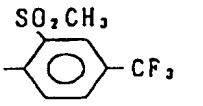
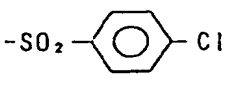
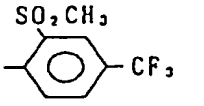
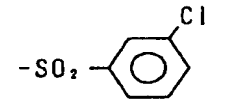
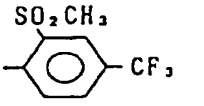
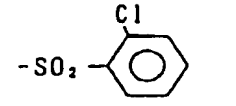
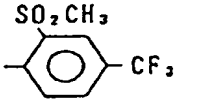
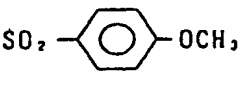
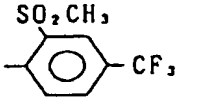
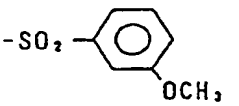
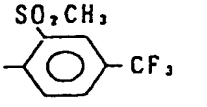
Compound No.	R_1	R_2		Physical properties
a − 37	CH_3			
a − 38	CH_3			
a − 39	CH_3	$-SO_2CH_3$		m.p. 143 − 146°C
a − 40	CH_3	$-SO_2CH_2CH_3$		m.p. 127 − 130.5°C
a − 41	CH_3			m.p. 117 − 120°C
a − 42	CH_3			m.p. 196 − 199°C
a − 43	CH_3			m.p. 157 − 160°C
a − 44	CH_3			m.p. 168 − 171°C
a − 45	CH_3			m.p. 157 − 160°C
a − 46	CH_3			

Table 4a (continued)

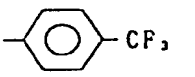
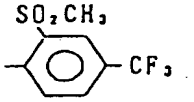
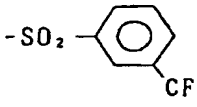
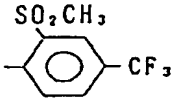
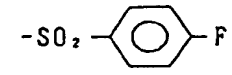
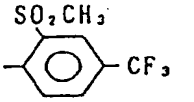
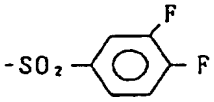
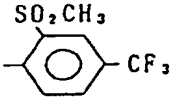
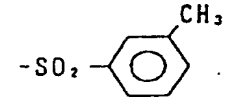
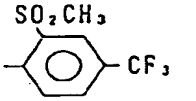
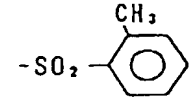
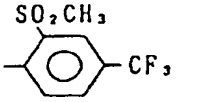
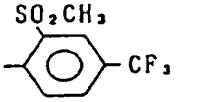
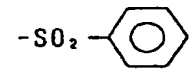
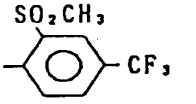
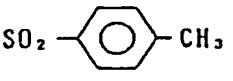
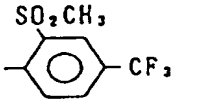
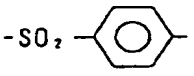
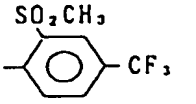
Compound No.	R_1	R_2	(X)n	Physical properties
a-47	CH_3			m.p. 185 - 188°C
a-48	CH_3			m.p. 124 - 127°C
a-49	CH_3			m.p. 152 - 155°C
a-50	CH_3			
a-51	CH_3			m.p. 166 - 169°C
a-52	CH_3			m.p. 145 - 149°C
a-53	CH_3	$-SO_2C_4H_9(n)$		m.p. 142 - 145°C
a-54	CH_2CH_3			
a-55	CH_2CH_3			m.p. 156 - 159°C
a-56	CH_2CH_3			

Table 4a (continued)

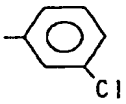
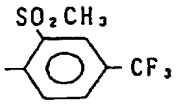
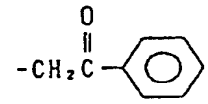
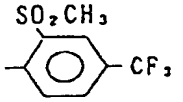
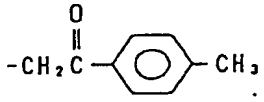
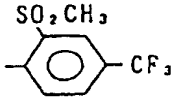
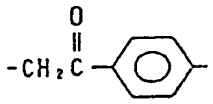
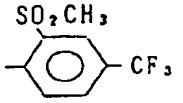
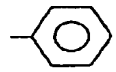
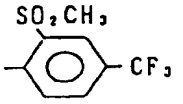
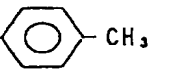
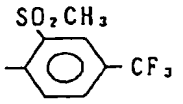
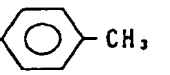
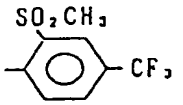
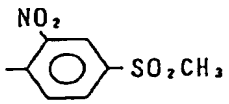
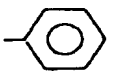
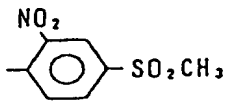
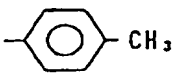
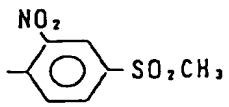
Compound No.	R_1	R_2	(X)n	Physical properties
a − 57	CH_2CH_3			
a − 58	CH_2CH_3			m.p. 136 - 138°C
a − 59	CH_3			Viscous
a − 60	CH_3			Viscous
a − 61	$CH(CH_3)_2$			
a − 62	$CH(CH_3)_2$			
a − 63	$n\text{-}C_3H_7$			
a − 64	CH_3	$-SO_2CH_3$		
a − 65	CH_3			
a − 66	CH_3			

Table 4a (continued)

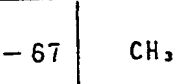
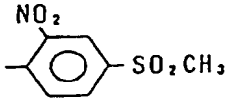
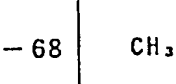
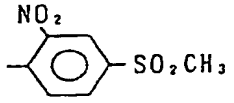
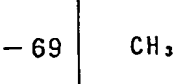
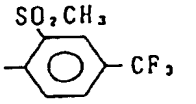
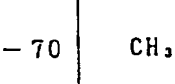
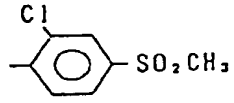
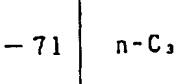
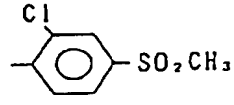
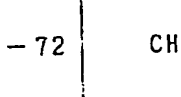
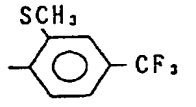
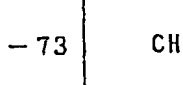
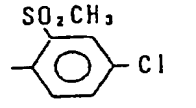
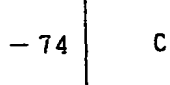
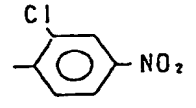
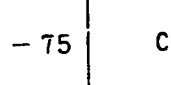
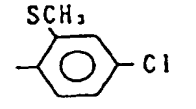
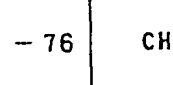
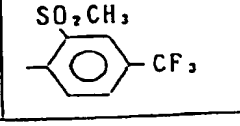
Compound No.	R_1	R_2	(X)n	Physical properties
a-67	CH_3			
a-68	CH_3			
a-69	CH_3			m.p. 208-211℃
a-70	CH_3			
a-71	$n\text{-}C_3H_7$			
a-72	CH_3			m.p. 107-109℃
a-73	CH_3			m.p. 158-164℃
a-74	CH_3			
a-75	CH_3			
a-76	CH_3			Viscous

Table 4a (continued)

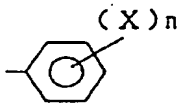
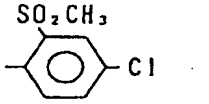
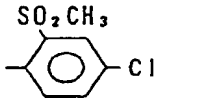
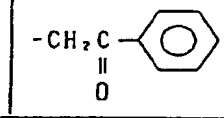
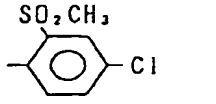
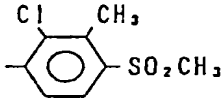
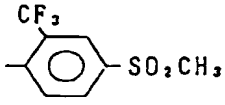
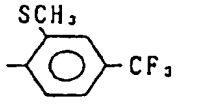
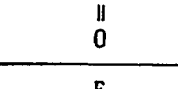
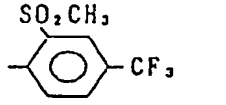
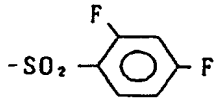
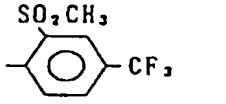
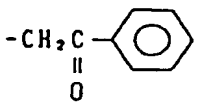
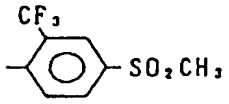
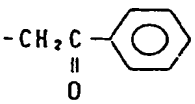
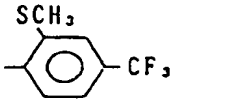
Compound No.	R_1	R_2		Physical properties
a－77	CH_3	H		m.p. 190－204℃
a－78	CH_3	$-SO_2CH_3$		m.p. 134－138℃
a－79	CH_3			m.p. 137－139℃
a－80	CH_3	H		m.p. 177－180℃
a－81	CH_3	H		m.p. 141－143℃
a－82	CH_3	H		m.p. 107－110℃
a－83	CH_3			Viscous
a－84	CH_3			m.p. 189－193℃
a－85	CH_3			m.p. 113－115℃
a－86	CH_3			m.p. 146－148℃

Table 4a (continued)

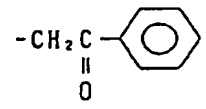
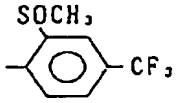
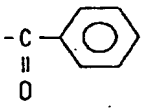
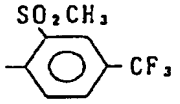
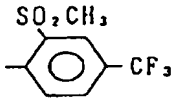
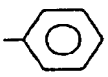
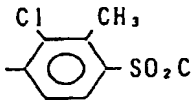
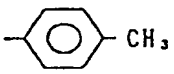
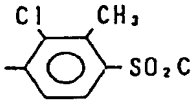
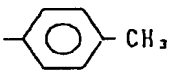
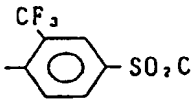
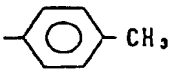
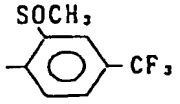
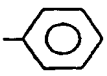
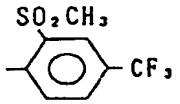
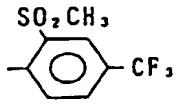
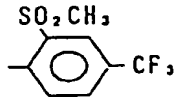
Compound No.	R_1	R_2	(X)n	Physical properties
a-87	CH_3			m.p. 148-151°C
a-88	CH_3			m.p. 137-141°C
a-89	CH_3	$-SO_2C_3H_7(n)$		m.p. 128-131°C
a-90	CH_3			Viscous
a-91	CH_3			m.p. 188-192°C
a-92	CH_3			Viscous
a-93	CH_3			m.p. 128-131°C
a-94	CH_3			m.p. 154-157°C
a-95	CH_3	$-CH_2CN$		m.p. 135-140°C
a-96	CH_3	$-CH_2CH=CH_2$		Refractive index $n_D^{44.5}$ 1.5133

Table 4a (continued)

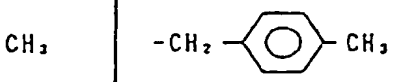
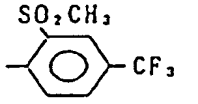
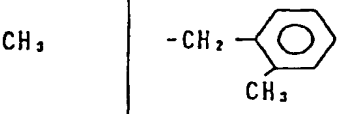
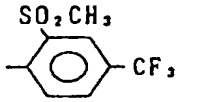
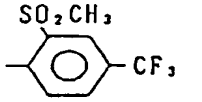
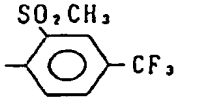
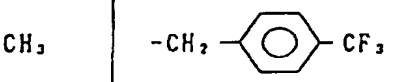
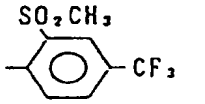
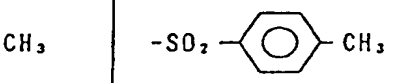
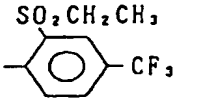
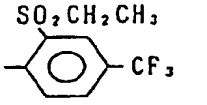
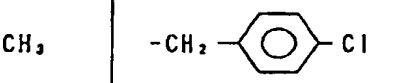
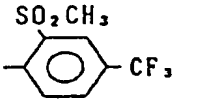
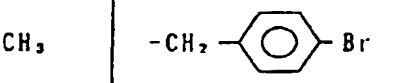
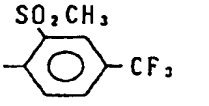
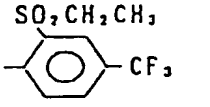
Compound No.	R_1	R_2	(X)n	Physical properties
a-97	CH_3			m.p. 161-163°C
a-98	CH_3			m.p. 163-166°C
a-99	CH_3	$-CH_2C\equiv CH$		m.p. 123-127°C
a-100	CH_3	$-CH_2CH_3$		m.p. 125-128°C
a-101	CH_3			m.p. 159-161°C
a-102	CH_3			m.p. 63-70°C
a-103	CH_3	$-SO_2C_3H_7(n)$		m.p. 130-133°C
a-104	CH_3			m.p. 151-154°C
a-105	CH_3			m.p. 160-163°C
a-106	CH_3	H		m.p. 140-143°C

Table 4a (continued)

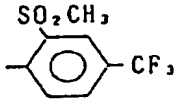
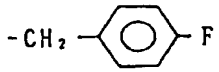
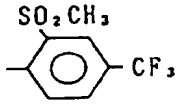
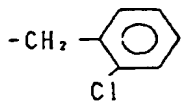
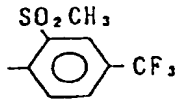
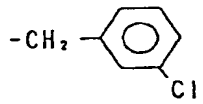
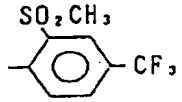
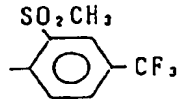
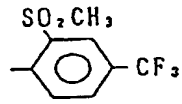
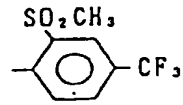
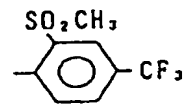
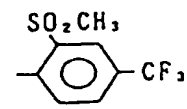
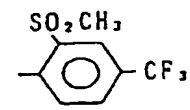
Compound No.	R_1	R_2	(X)n	Physical properties
a-107	CH_2CH_3	H		m.p. 109-114°C
a-108	CH_3			
a-109	CH_3			
a-110	CH_3			
a-111	CH_3	$-CH_2OCH_3$		
a-112	CH_3	$-CH_2OCH_2CH_3$		
a-113	CH_3	$-CH_2\overset{O}{\overset{\|}{C}}OCH_3$		Viscous
a-114	CH_3	$-CH_2\overset{O}{\overset{\|}{C}}OCH_2CH_3$		
a-115	CH_3	$-CH_3$		
a-116	CH_3	$-C_3H_7(n)$		Viscous

Table 4a (continued)

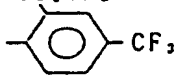
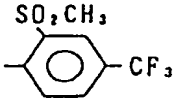
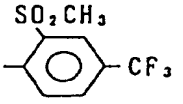
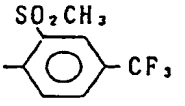
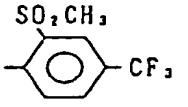
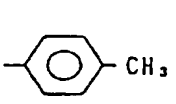
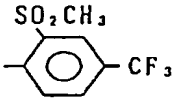
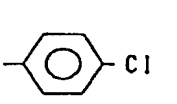
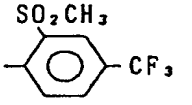
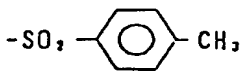
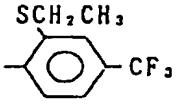
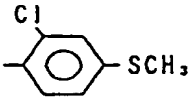
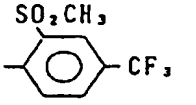
Compound No.	R_1	R_2	(X)n	Physical properties
a-117	CH_3	$-C_4H_9(n)$		
a-118	CH_3	$-\overset{O}{\overset{\|}{C}}CH_3$		m.p. 156 - 158°C
a-119	CH_3	$-\overset{O}{\overset{\|}{C}}CH_2CH_3$		
a-120	CH_3	$-\overset{O}{\overset{\|}{C}}-C_3H_7(n)$		m.p. 143 - 145°C
a-121	CH_3	$-\overset{O}{\overset{\|}{C}}-C_4H_9(n)$		
a-122	CH_3			m.p. 179 - 182°C
a-123	CH_3			
a-124	CH_3			
a-125	CH_3	$-SO_2N(CH_3)_2$		m.p. 116 - 118°C
a-126	CH_3	$-SO_2N(CH_3)_2$		m.p. 154 - 158°C

Table 4a (continued)

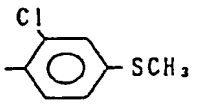
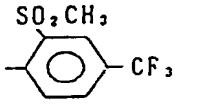
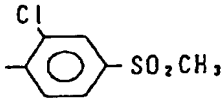
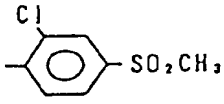
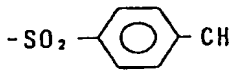
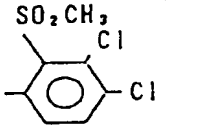
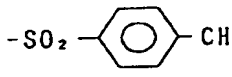
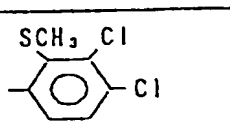
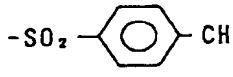
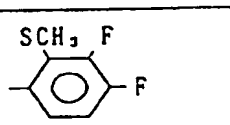
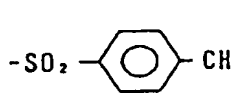
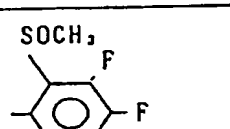
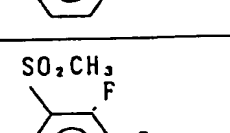
Compound No.	R_1	R_2	(X)n	Physical properties
a-127	CH_3	$-CON(CH_3)_2$		m.p. 136 - 138°C
a-128	CH_3	$-CON(CH_3)_2$		
a-129	CH_3	$-SO_2N(CH_3)_2$		m.p. 50 - 60°C
a-130	CH_3	$-CON(CH_3)_2$		m.p. 200 - 203°C
a-131	CH_3			m.p. 57 - 60°C
a-132	CH_3			Viscous
a-133	CH_3			m.p. 105 - 107°C
a-134	CH_3			m.p. 131 - 133°C
a-135	CH_3			m.p. 169 - 172°C

Table 4a (continued)

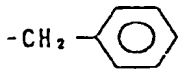
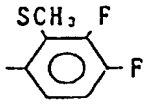
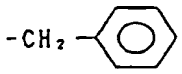
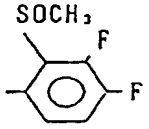
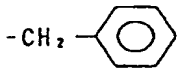
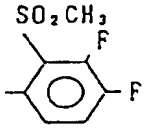
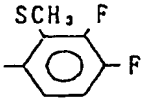
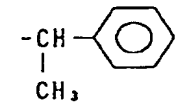
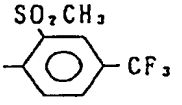
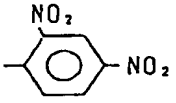
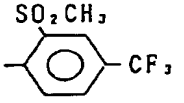
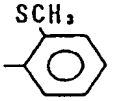
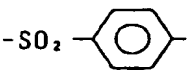
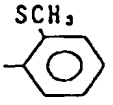
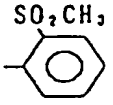
Compound No.	R_1	R_2	(X)n	Physical properties
a-136	CH_3			Oily
a-137	CH_3			Viscous
a-138	CH_3			m.p. 125-128°C
a-139	CH_3	H		Oily
a-140	CH_3			m.p. 139-142°C
a-141	CH_3			m.p. 150-151°C
a-142	CH_3	H		m.p. 95-103°C
a-143	CH_3			m.p. 92-96°C
a-144	CH_3	H		m.p. 60-70°C

Table 4a (continued)

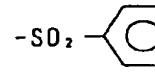
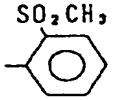
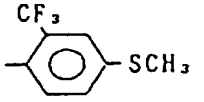
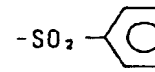
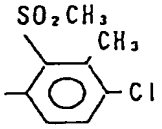
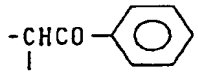
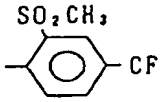
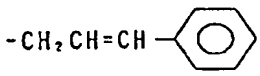
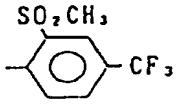
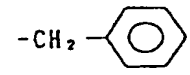
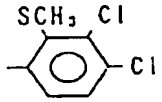
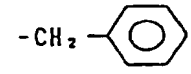
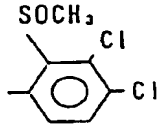
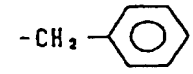
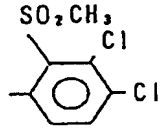
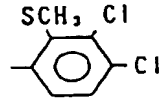
Compound No.	R_1	R_2	(X)n	Physical properties
a-145	CH_3			m.p. 75 - 80°C
a-146	CH_3	H		m.p. 70 - 85°C
a-147	CH_3			
a-148	CH_3			m.p. 152 - 153°C
a-149	CH_3			Viscous
a-150	CH_3			Viscous
a-151	CH_3			Viscous
a-152	CH_3			m.p. 157 - 160°C
a-153	CH_3	$-SO_2C_3H_7(n)$		m.p. 82 - 85°C

Table 4a (continued)

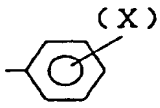
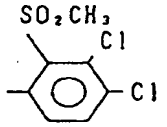
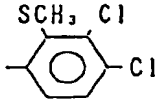
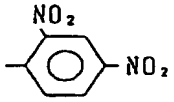
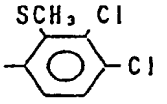
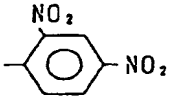
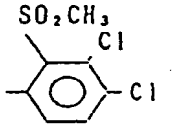
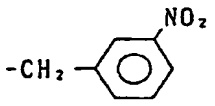
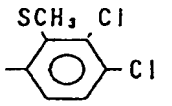
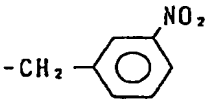
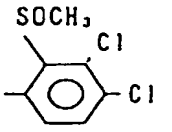
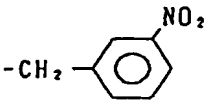
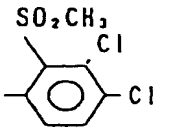
Compound No.	R_1	R_2		Physical properties
a-154	CH_3	$-SO_2C_3H_7(n)$		Viscous
a-155	CH_3	$-SO_2C_3H_7(n)$		m.p. 165-169°C
a-156	CH_3	H		Viscous
a-157	CH_3			Viscous
a-158	CH_3			m.p. 120-130°C
a-159	CH_3			Viscous
a-160	CH_3			m.p. 177-178°C
a-161	CH_3			m.p. 173-175°C

Table 4a (continued)

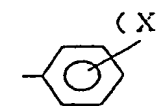
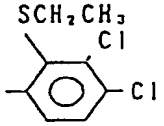
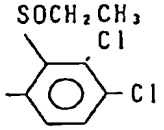
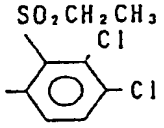
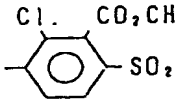
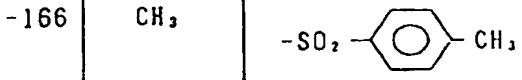
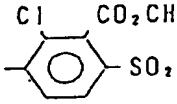
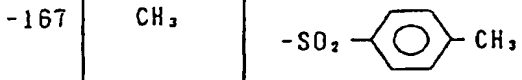
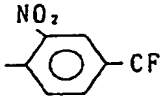
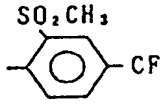
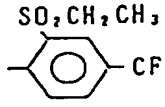
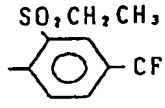
Compound No.	R_1	R_2		Physical properties
a-162	CH_3			Viscous
a-163	CH_3			Viscous
a-164	CH_3			m.p. 65 - 75°C
a-165	CH_3	H		m.p. 208°C (decomposition)
a-166	CH_3			m.p. 144 - 147°C
a-167	CH_3			m.p. 120 - 141°C
a-168	CH_3	$-CH_2CO-C(CH_3)_3$		m.p. 140 - 144°C
a-169	CH_3			m.p. 119 - 122°C
a-170	CH_3	$-CH_2C\equiv CH$		Oily

Table 4a (continued)

Compound No.	R_1	R_2	(X)n	Physical properties
a-171	CH_3	$-CH_2$-Ph	OSO_2CH_3, Cl	Viscous
a-172	CH_3	H	CH_2SCH_3, Cl	Oily
a-173	CH_3	$-CH_2CN$	$SOCH_3$, Cl, Cl	Viscous
a-174	CH_3	$-CH_2CN$	SO_2CH_3, Cl, Cl	Viscous
a-175	CH_3	$-SO_2$-Ph-CH_3	SCH_3, NO_2	m.p. 153 - 156°C
a-176	CH_3	$-SO_2$-Ph-CH_3	SO_2CH_3, NO_2	Viscous
a-177	CH_3	$-SO_2$-Ph-CH_3	SCH_3, Cl, CF_3	Viscous
a-178	CH_3	$-SO_2$-Ph-CH_3	SO_2CH_3, Cl, CF_3	Viscous
a-179	CH_3	$-SO_2C_3H_7$(n)	SO_2CH_3, Cl, CF_3	m.p. 140 - 144°C

Table 4a (continued)

Compound No.	R_1	R_2	(X)n	Physical properties
a-180	CH_3	$-CH_2-C_6H_5$	CH_2SCH_3, Cl	Viscous
a-181	CH_3	$-CH_2-C_6H_5$	CH_2SOCH_3, Cl	Viscous
a-182	CH_3	$-CH_2-C_6H_5$	$CH_2SO_2CH_3$, Cl	m.p. 55 - 62°C
a-183	CH_3	H	SCH_3, Cl, Cl	Viscous
a-184	CH_3	$-CH_2-C_6H_5$	SCH_3, Cl, Cl	Viscous
a-185	CH_3	$-CH_2-C_6H_5$	$SOCH_3$, Cl, Cl	m.p. 154 - 155°C
a-186	CH_3	$-CH_2-C_6H_5$	SO_2CH_3, Cl, Cl	m.p. 165 - 167°C
a-187	CH_3	H	$N(CH_3)SO_2CH_3$, Cl	m.p. 50 - 58°C

Table 4a (continued)

Compound No.	R_1	R_2	(X)n	Physical properties
a-188	CH_3	$-SO_2C_3H_7(n)$	$N(CH_3)SO_2CH_3$, Cl	m.p. 181-184°C
a-189	CH_3	$-SO_2$-Ph-CH_3	$N(CH_3)SO_2CH_3$, Cl	m.p. 70 - 73°C
a-190	CH_3	$-CH_2C\equiv CH$	SCH_3, Cl, Cl	Viscous
a-191	CH_3	$-CH_2C\equiv CH$	SO_2CH_3, Cl, Cl	m.p. 40 - 50°C
a-192	CH_3	H	SCH_3, CF_3, Cl	Viscous
a-193	CH_3	$-CH_2$-Ph	SCH_3, CF_3, Cl	m.p. 107 - 110°C
a-194	CH_3	$-CH_2$-Ph	$SOCH_3$, CF_3, Cl	m.p. 48 - 52°C
a-195	CH_3	$-CH_2$-Ph	SO_2CH_3, CF_3, Cl	m.p. 140 - 148°C
a-196	CH_3	H	SCH_3, CH_3	m.p. 115 - 130°C

Table 4a (continued)

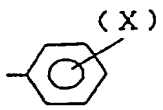
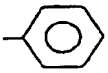
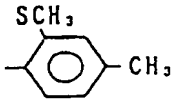
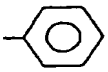
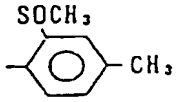
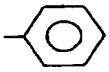
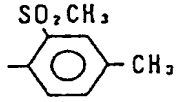
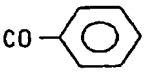
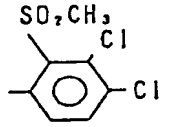
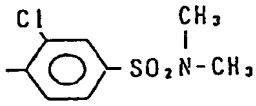
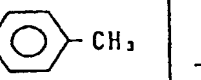
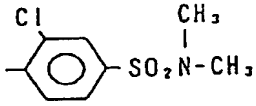
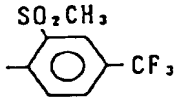
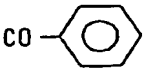
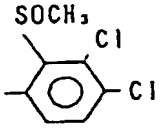
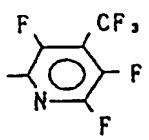
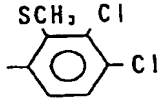
Compound No.	R_1	R_2		Physical properties
a-197	CH_3			m.p. 79 - 93°C
a-198	CH_3			m.p. 114 - 125°C
a-199	CH_3			m.p. 143 - 146°C
a-200	CH_3			m.p. 173 - 178°C
a-201	CH_3	H		m.p. 155 - 156°C
a-202	CH_3			m.p. 150 - 151°C
a-203	CH_3	$-SO_2CH_2CH(CH_3)_2$		m.p. 183 - 188°C
a-204	CH_3			Viscous
a-205	CH_3			m.p. 111 - 126°C

Table 4a (continued)

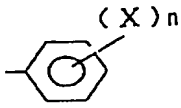
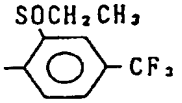
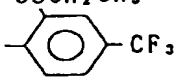
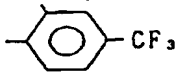
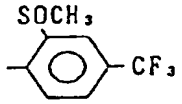
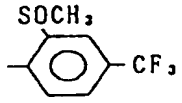
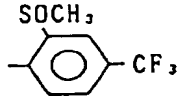
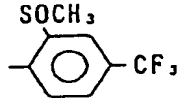
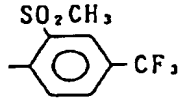
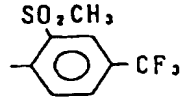
Compound No.	R_1	R_2		Physical properties
a -206	CH_3	$-CH_2C\equiv CH$		Oily
a -207	CH_3	$-SO_2C_3H_7(n)$		Viscous
a -208	CH_3	$-SO_2C_3H_7(n)$		m.p. 134 - 135°C
a -209	CH_3	$-CH_2C\equiv CH$		Viscous
a -210	CH_3			Viscous
a -211	CH_3	H		m.p. 122 - 138°C
a -212	CH_3	$-CH_2CN$		Viscous
a -213	CH_3	$-CH_2C(Cl)=CH_2$		m.p. 108 - 111°C
a -214	CH_3	$-SO_2(CH_2)_3Cl$		m.p. 113 - 118°C

Table 4a (continued)

Compound No.	R_1	R_2	(X)n	Physical properties
a -215	CH_3	$-CH_2$-Ph	$CH_2SO_2CH_3$, Cl, Cl	m.p. 50 – 60°C
a -216	CH_3	$-CO(CH_2)_3Cl$	SO_2CH_3, CF_3	m.p. 133 – 135°C
a -217	CH_3	$-CH_2CH=CH_2$	$SOCH_2CH_3$, CF_3	Viscous
a -218	CH_3	$-CH_2CN$	$SOCH_2CH_3$, CF_3	Viscous
a -219	CH_3	$-CH_2$-Ph	$SOCH_2CH_3$, CF_3	Viscous
a -220	CH_3	$-SO_2CH_3$	$SOCH_2CH_3$, CF_3	m.p. 125 – 130°C
a -221	CH_3	$-CO_2CH_3$	$SOCH_2CH_3$, CF_3	Viscous
a -222	CH_3	H	$SOCH_2CH_3$, CF_3	m.p. 153 – 156°C
a -223	CH_3	$-CH_2CO_2CH_2CH_3$	$SOCH_2CH_3$, CF_3	Viscous

Table 4a (continued)

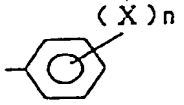
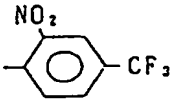
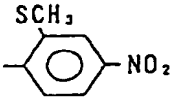
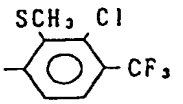
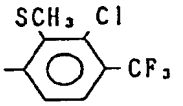
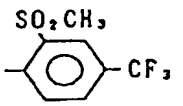
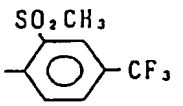
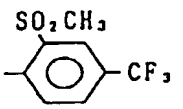
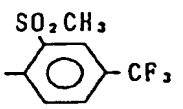
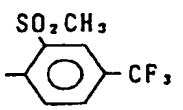
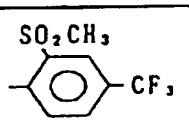
Compound No.	R_1	R_2		Physical properties
a -224	CH_3	H		
a -225	CH_3	H		Viscous
a -226	CH_3	$-SO_2C_3H_7(n)$		Viscous
a -227	CH_3	H		Oily
a -228	CH_3	$-CH_2CH=CHCl$		m.p. 100 - 103°C
a -229	CH_3	$-CH_2C(CH_3)=CH_2$		m.p. 122 - 125°C
a -230	CH_3	$-CH_2C(Br)=CH_2$		Oily
a -231	CH_3	$-CH_2CH=C(CH_3)_2$		
a -232	CH_3	$-CH_2CH=C(Cl)_2$		m.p. 142 - 145°C
a -233	CH_3	$-CH_2CH=CHCH_3$		m.p. 101 - 106°C

Table 4a (continued)

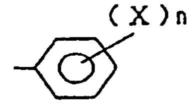
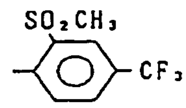
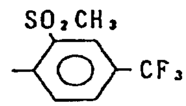
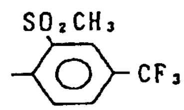
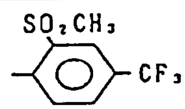
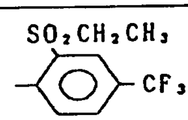
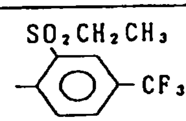
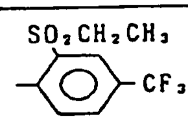
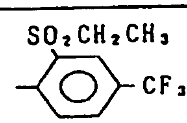
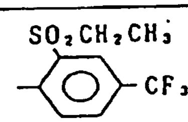
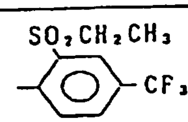
Compound No.	R_1	R_2		Physical properties
a -234	CH_3	$-CHCH=CH_2$ | CH_3		Oily
a -235	CH_3	$-CH_2CON(CH_3)_2$		
a -236	CH_2CH_3	$-CH_2C=CH_2$ | Cl		
a -237	CH_2CH_3	$-CH_2C=CH_2$ | CH_3		
a -238	CH_3	$-CH_2C=CH_2$ | CH_3		
a -239	CH_3	$-CH_2C=CH_2$ | Br		m.p. 124 - 126°C
a -240	CH_3	$-CH_2CH=C(CH_3)_2$		
a -241	CH_3	$-CHCH=CH_2$ | CH_3		Viscous
a -242	CH_3	$-SO_2CH_2CH_3$		
a -243	CH_3	$-SO_2C_4H_9(n)$		

Table 4a (continued)

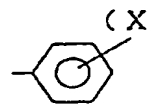
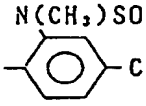
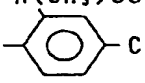
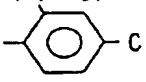
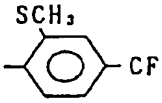
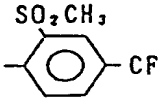
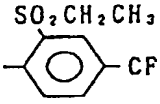
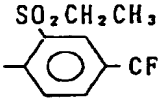
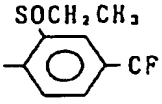
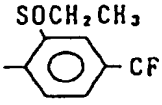
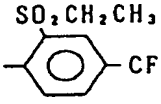
Compound No.	R_1	R_2		Physical properties
a -244	CH_3	H		m.p. 60 - 65°C
a -245	CH_3	$-SO_2C_3H_7(n)$		
a -246	CH_3	$-CH_2C(Cl)=CH_2$		
a -247	CH_3	$-CH_2C(Cl)=CH_2$		
a -248	CH_3	$-CH_2C(CH_3)=CH_2$		
a -249	CH_3	$-CH_2CH=CH_2$		Viscous
a -250	CH_3	$-CH_2CN$		Viscous
a -251	CH_3	$-CH_2C(Cl)=CH_2$		Viscous
a -252	CH_3	$-CH_2CH=CHCl$		Viscous
a -253	CH_3	$-CH_2CH=CHCl$		Viscous

Table 4a (continued)

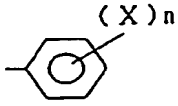
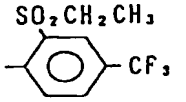
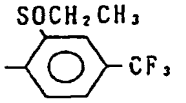
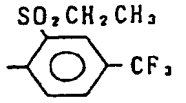
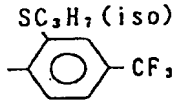
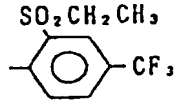
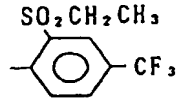
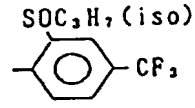
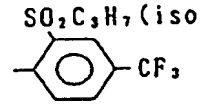
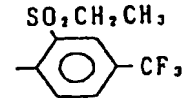
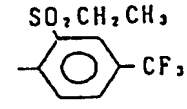
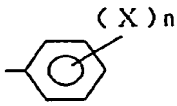
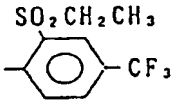
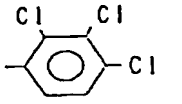
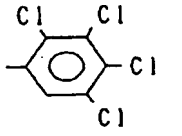
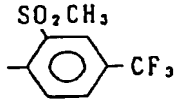
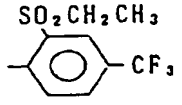
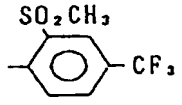
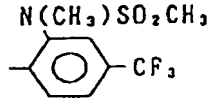
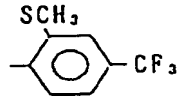
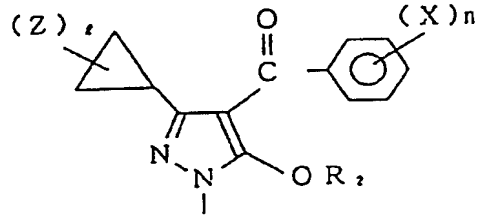
Compound No.	R_1	R_2		Physical properties
a -254	CH_3	$-CH_2C(Cl)=CH_2$		m.p. 120 - 125℃
a -255	CH_3	$-CH_2CH=CHCH_3$		m.p. 97 - 105℃
a -256	CH_3	$-CH_2CH=CHCH_3$		m.p. 116 - 117℃
a -257	CH_3	$-SO_2CH_2CH_3$		Viscous
a -258	CH_3	$-SO_2CH_3$		m.p. 164 - 165℃
a -259	CH_3	$-CH_2CO_2CH_2CH_3$		Viscous
a -260	CH_3	$-SO_2CH_2CH_3$		m.p. 112 - 115℃
a -261	CH_3	$-SO_2CH_2CH_3$		Viscous
a -262	CH_3	$-CH_2C(=O)CH_2C(=O)CH_2CH_3$		Viscous
a -263	CH_3	$-CH_2CON(C_2H_5)_2$		Viscous

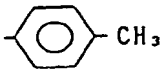
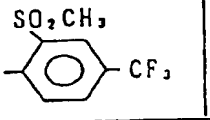
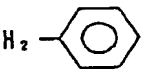
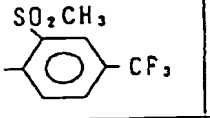
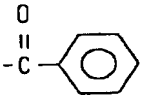
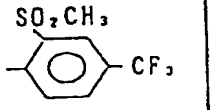
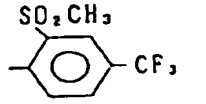
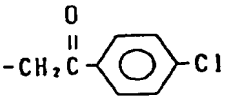
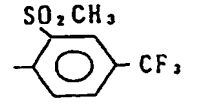
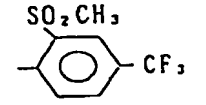
Table 4 (continued)

Compound No.	R_1	R_2		Physical properties
a -264	CH_3	$-CH_2CHCH_2Cl$ (CH$_3$ on CH)		m.p. 123～125℃
a -265	CH_3	$-SO_2C_3H_7(n)$		
a -266	CH_3	$-SO_2C_3H_7(n)$		
a -267	CH_3	$-CH_2CH_2Cl$		
a -268	CH_3	$-CH_2CH_2Cl$		
a -269	CH_3	$-COC=CH_2$ (CH$_3$ on C)		
a -270	CH_3	$-SO_2$-Ph-CH_3		m.p. 67～ 72℃
a -271	CH_3	$-CH_2CH=C(Cl)_2$		Viscous



... (I)

Table 4b

Comp. No.	R_1	(Z)t	R_2	(X)n	Physical properties
b-1	CH_3				m.p. 46-56°C
b-2	CH_3				Viscous
b-3	CH_3				
b-4	CH_3		$-SO_2C_3H_7(n)$		
b-5	CH_3				
b-6	CH_3		H		

Now, the Test Examples of the present invention will be described.

TEST EXAMPLE 1

Upland field soil was put into a 1/150,000ha pot, and seeds of various plants were sown. Then, when the plants reached predetermined leaf stages (① barnyardgrass (Echinochloa crus-galli L.), EC: 1.3-2.6 leaf stage, ② crabgrass (Digitaria sanguinalis L.), DS: 1.0-2.5 leaf stage, ③ redroot pigweed (Amaranthus retroflexus L.), AR: 0.1-1.2 leaf stage, ④ prickly sida (Sida spinosa L.), SS: 0.1-1.2 leaf stage, ⑤ tall morningglory (Pharbitis purpurea L.), PP: 0.3-1.3 leaf stage, ⑥ common cocklebur (Xanthium strumarium L.), XS: 0.1-1.8 leaf stage, ⑦ rice (Oryza sativa L.), OS: 1.0-2.5 leaf stage, ⑧ wheat (Triticum spp.), TR: 2.2-2.9 leaf stage, ⑨ corn (Zea mays L.), ZM: 1.8-3.5 leaf stage, ⑩ soybean (Glycine max Merr.), GM: primary leaf - 0.3 leaf stage), a wettable powder having the compound of the present invention formulated in accordance with a usual formulation method, was weighed so that the active ingredient would be a predetermined amount, and diluted with water in an amount of 500 ℓ/ha. To the diluted solution, 0.1 %(v/v) of an agricultural spreader was added. The herbicide thus adjusted was applied by a small size spray for foliage treatment. On the 18th to 30th days after the application of the herbicide, the growth of the respective plants was visually observed,

and the herbicidal effects were evaluated by the growth-controlling degrees (%) ranging from 0 (equivalent to the non-treated area) to 100 (complete kill), whereby the results shown in Table 5, were obtained. Compound Nos. in Table 5 correspond to Compound Nos. in Table 4a and 4b given hereinbefore.

Table 5

Comp. No.	Dose of active ingredient (g/ha)	Growth-controlling degree (%)										Evaluation day
		EC	DS	AR	SS	PP	XS	OS	TR	ZM	GM	
a-3	500	0	0	70	60	60	70	40	0	0	0	23
a-5	500	30	40	70	0	60	60	50	10	0	10	18
a-11	63	40	20	90	20	80	80	50	0	0	50	22
	125	70	40	90	20	80	100	80	0	0	70	
	500	90	50	100	50	100	100	100	0	0	80	
a-12	63	80	40	90	30	80	95	80	0	20	70	18
	125	80	70	80	20	90	80	90	0	0	70	
	500	90	80	90	60	90	80	100	0	0	80	
a-13	500	80	60	90	60	80	100	90	0	0	60	18
a-24	500	40	50	80	10	60	80	50	-	10	30	18
a-25	500	70	90	80	10	70	90	50	-	30	60	18
a-27	500	60	20	100	20	80	80	50	0	0	40	20
a-39	63	70	30	80	20	60	80	50	0	10	60	20
	125	80	60	90	30	60	100	70	0	10	60	
	500	90	80	90	30	70	100	70	10	20	80	
a-40	63	90	30	90	10	90	80	90	-	30	50	18
	125	90	40	90	30	90	90	100	-	50	70	
	500	100	90	100	60	90	100	100	-	70	70	
a-41	63	90	20	90	20	90	70	90	-	0	50	18
	125	90	40	90	20	90	80	90	-	0	70	
	500	100	90	100	40	100	100	100	-	20	90	
a-42	500	60	40	100	30	70	80	50	10	20	50	20
a-43	125	50	30	90	20	60	80	40	0	20	50	20
	500	70	80	100	30	80	100	70	20	20	70	
a-44	63	80	50	80	20	60	80	60	-	0	60	18
	125	90	70	80	20	70	90	80	-	20	70	
	500	90	90	90	30	90	100	80	-	20	80	
a-45	125	40	30	80	20	80	60	10	10	40	50	20
	500	70	50	90	20	80	100	20	20	20	50	
a-47	125	40	40	90	30	60	80	20	-	0	0	18
	500	80	50	100	60	80	80	70	-	30	40	

Table 5 (continued)

Comp. NO.	Dose of active ingredient (g/ha)	Growth-controlling degree (%)										Evaluation day
		EC	DS	AR	SS	PP	XS	OS	TR	ZM	GM	
a-48	63	80	40	90	10	40	70	50	-	0	50	18
	125	90	60	90	30	70	100	60	-	20	60	
	500	100	80	100	60	90	100	80	-	40	70	
a-49	125	80	40	100	30	70	90	60	10	20	60	20
	500	80	70	100	30	80	90	80	20	20	60	
a-51	125	90	60	80	10	80	80	70	-	0	70	19
	500	90	90	100	30	90	90	80	-	10	70	
a-52	63	90	60	80	20	70	80	60	-	20	80	18
	125	90	70	80	30	80	90	70	-	30	90	
	500	90	90	90	50	90	100	80	-	40	90	
a-53	63	70	10	90	0	70	80	50	-	0	50	18
	125	80	20	90	10	80	90	50	-	0	60	
	500	90	90	90	20	90	100	90	-	10	70	
a-59	500	70	40	80	0	80	90	30	0	10	60	20
a-60	63	90	50	80	10	90	90	60	-	0	100	19
	125	90	80	90	20	100	100	70	-	20	90	
	500	90	90	100	30	100	100	90	-	60	90	
a-69	500	40	40	90	50	70	100	60	0	10	40	20
a-72	125	70	20	80	0	60	90	50	-	0	40	19
	500	100	30	90	0	70	100	80	-	0	60	
a-73	500	60	30	90	40	70	80	10	-	30	50	18
a-76	63	70	30	90	0	80	90	70	0	10	70	20
	125	80	60	90	20	80	90	70	10	20	70	
	500	90	90	100	40	100	100	70	20	40	80	
a-77	125	90	30	80	10	70	70	50	-	10	60	18
	500	90	60	90	30	70	90	90	-	30	70	
a-78	63	80	40	70	20	60	80	40	-	10	60	18
	125	80	40	90	20	90	90	60	-	20	70	
	500	100	50	100	40	90	100	60	-	50	70	
a-79	500	60	30	100	50	80	90	30	-	0	70	18
a-80	500	80	90	80	20	60	80	80	-	60	10	19

Table 5 (continued)

Comp. No.	Dose of active ingredient (g/ha)	Growth-controlling degree (%)										Evaluation day
		EC	DS	AR	SS	PP	XS	OS	TR	ZM	GM	
a-81	125	60	60	90	10	90	40	10	-	0	30	19
	500	90	90	90	30	90	-	40	-	10	50	
a-82	125	40	30	80	20	60	100	60	-	0	60	19
	500	80	30	90	20	90	100	80	-	10	70	
a-83	63	50	60	80	30	70	100	60	-	0	10	18
	125	60	70	80	40	80	100	60	-	10	20	
	500	90	90	90	40	90	90	80	-	30	70	
a-84	63	70	30	70	30	40	100	30	-	0	50	18
	125	80	50	80	20	60	100	60	-	0	60	
	500	90	90	90	50	90	90	100	-	20	70	
a-87	125	80	20	90	10	70	100	60	-	10	60	19
	500	100	70	95	10	100	-	80	-	0	95	
a-88	63	90	60	80	30	80	90	70	-	10	70	18
	125	90	80	90	60	80	90	70	-	20	80	
	500	100	90	90	60	90	100	80	-	60	90	
a-89	63	90	90	90	30	90	100	100	-	10	90	19
	125	90	90	90	30	90	100	90	-	20	100	
	500	100	100	100	60	100	100	100	-	80	100	
a-90	125	80	90	70	0	60	80	60	-	40	10	19
	500	90	100	80	10	70	100	80	-	80	70	
a-92	500	80	90	90	20	90	-	50	-	0	20	19
a-93	125	60	40	95	0	60	100	10	-	0	70	19
	500	80	50	100	20	100	-	70	-	0	100	
a-94	63	90	90	80	10	90	80	70	-	0	60	19
	125	100	90	90	-	100	100	90	-	10	70	
	500	90	90	90	20	100	100	90	-	20	70	
a-95	63	100	70	100	10	80	90	90	-	20	95	19
	125	100	90	100	10	95	100	100	-	35	95	
	500	100	100	100	40	100	-	100	-	70	100	
a-96	125	80	70	80	20	80	90	60	10	40	80	20
	500	100	100	100	30	100	100	70	20	60	90	
a-97	63	90	30	90	0	40	50	10	-	0	20	18
	125	80	40	80	10	50	70	30	-	0	30	
	500	90	90	90	20	70	80	50	-	20	60	

Table 5 (continued)

Comp. No.	Dose of active ingredient (g/ha)	Growth-controlling degree (%)										Evaluation day
		EC	DS	AR	SS	PP	XS	OS	TR	ZM	GM	
a-98	63	90	30	90	0	60	70	30	-	0	40	
	125	90	40	100	30	80	70	40	-	0	50	18
	500	90	90	90	30	90	100	100	-	20	60	
a-99	63	90	40	100	40	80	80	70	-	40	70	
	125	100	60	100	40	90	100	90	-	50	70	18
	500	100	100	100	70	100	100	100	-	80	100	
a-100	125	80	40	90	30	80	70	0	-	0	50	18
	500	90	90	100	40	90	100	50	-	10	60	
a-101	125	90	30	90	0	30	20	0	-	0	50	18
	500	90	40	90	30	30	60	0	-	20	60	
a-102	63	90	40	90	20	70	80	80	-	0	100	
	125	95	60	90	30	80	80	100	-	10	60	19
	500	100	90	100	70	90	80	90	-	20	100	
a-103	63	90	30	90	40	60	70	50	-	0	50	
	125	100	50	60	40	80	100	50	-	0	50	19
	500	100	90	80	50	100	100	100	-	40	70	
a-104	125	80	20	90	0	10	0	0	-	0	10	19
	500	90	20	100	10	10	0	0	-	0	20	
a-105	63	90	30	100	30	40	20	10	-	10	40	
	125	90	40	100	0	60	80	30	-	0	50	19
	500	100	80	80	30	90	-	50	-	80	90	
a-106	63	10	30	80	50	60	-	20	-	0	30	
	125	70	70	100	50	90	90	90	-	20	40	21
	500	90	70	70	-	100	50	100	-	50	80	
a-116	125	50	10	100	30	70	-	30	-	10	30	24
	500	50	50	90	20	90	-	20	-	0	40	
a-118	63	70	30	100	30	70	90	100	-	10	70	
	125	80	50	80	30	80	-	100	-	40	100	21
	500	90	80	100	-	100	100	100	-	60	100	
a-120	63	90	50	100	20	50	80	90	-	0	70	
	125	100	50	100	50	80	100	100	-	0	80	21
	500	90	90	-	100	60	100	100	-	70	100	
a-122	63	70	50	100	40	60	90	60	-	10	50	
	125	80	90	100	50	70	90	90	-	30	60	21
	500	90	90	-	-	50	100	100	-	40	70	

Table 5 (continued)

Comp. No.	Dose of active ingredient (g/ha)	Growth-controlling degree (%)										Evaluation day
		EC	DS	AR	SS	PP	XS	OS	TR	ZM	GM	
a-131	500	80	40	60	30	80	100	50	-	30	20	19
a-132	125 500	80 90	70 90	60 60	20 20	90 90	50 50	10 40	- -	30 70	0 10	19
a-133	500	10	30	90	40	70	10	0	-	40	30	21
a-136	125 500	20 100	30 100	90 80	50 30	70 70	- 50	0 0	- -	40 100	40 50	21
a-137	125 500	20 90	30 60	90 100	0 10	100 100	- 80	0 40	- -	40 80	30 50	21
a-138	500	100	80	90	70	100	20	0	-	90	50	21
a-139	500	10	50	90	50	70	70	0	-	80	50	21
a-140	63 125 500	90 90 100	10 30 90	100 100 -	10 20 30	40 80 100	70 80 80	0 20 90	- - -	0 10 0	30 40 50	21
a-141	125 500	80 90	60 90	100 -	30 60	90 100	90 100	90 90	- -	20 30	70 80	21
a-143	500	90	30	50	20	80	100	20	-	20	60	18
a-144	500	50	90	30	40	60	100	50	-	0	20	18
a-146	125 500	20 60	30 80	80 80	40 40	80 90	50 100	40 60	- -	20 30	40 70	19
a-148	125 500	60 90	10 80	100 100	50 50	70 80	10 20	20 20	- -	10 40	10 40	21
a-149	63 125 500	60 90 100	20 30 80	90 80 100	40 50 70	80 80 100	- - 100	0 40 100	- - -	10 20 60	60 70 90	21
a-150	125 500	90 100	70 90	50 70	20 20	80 100	- 70	40 70	- -	100 100	60 70	25

Table 5 (continued)

Comp. No.	Dose of active ingredient (g/ha)	Growth-controlling degree (%)										Evaluation day
		EC	DS	AR	SS	PP	XS	OS	TR	ZM	GM	
a-151	125 500	100 100	90 90	40 100	30 30	80 100	30 80	20 20	- -	50 90	30 50	25
a-152	125 500	100 100	60 90	70 90	10 50	60 100	20 100	10 20	- -	60 50	10 40	25
a-153	125 500	60 90	20 60	50 60	10 30	40 50	80 -	10 20	- -	0 10	0 20	24
a-154	500	90	70	70	10	70	60	50	-	0	30	25
a-155	500	60	70	100	10	70	100	20	-	0	40	25
a-156	250	90	70	20	20	70	-	20	-	10	40	21
a-158	125 250	60 70	40 50	90 90	20 20	70 90	70 -	50 50	- -	40 60	50 50	21
a-162	500	60	20	0	10	80	-	10	0	10	0	21
a-164	500	70	70	20	10	90	-	10	0	40	50	21
a-165	63 125 500	100 100 100	95 95 100	100 100 100	40 100 100	100 100 100	50 - -	99 100 100	50 50 80	50 90 95	40 60 100	30
a-166	63 125 500	95 100 100	80 95 100	100 - 100	40 50 50	90 100 100	20 20 -	70 100 100	10 50 50	40 40 70	40 40 50	30
a-167	125 500	10 40	20 30	100 100	0 90	100 100	90 100	40 70	- -	0 30	40 100	21
a-168	63 125 500	90 100 100	60 70 100	100 100 100	50 60 60	90 100 100	90 - -	100 100 100	- - -	40 70 100	50 70 100	21
a-169	63 125	90 90	60 70	100 90	30 40	100 100	10 10	10 20	- -	10 10	30 60	21
a-170	63 125	90 90	60 80	80 100	20 50	100 100	70 70	90 100	- -	10 60	90 100	21

Table 5 (continued)

Comp. No.	Dose of active ingredient (g/ha)	Growth-controlling degree (%)										Evaluation day
		EC	DS	AR	SS	PP	XS	OS	TR	ZM	GM	
a-171	500	40	10	40	0	80	-	0	0	0	60	21
a-172	125	60	20	80	30	80	80	80	0	0	50	21
	500	100	50	90	60	100	-	100	0	50	95	
a-173	125	70	30	30	10	80	30	100	0	0	50	21
	500	90	60	40	30	100	-	100	0	40	60	
a-174	125	70	40	50	0	80	70	100	0	0	70	21
	500	100	60	90	30	100	-	80	0	50	80	
a-177	500	80	50	30	0	90	-	10	0	20	40	21
a-178	500	60	40	60	0	80	-	50	0	0	50	21
a-179	500	70	70	90	90	70	-	50	0	0	80	21
a-180	125	80	20	50	20	100	50	100	0	0	70	21
	500	100	50	90	30	100	-	50	0	60	80	
a-181	125	70	20	70	10	100	70	50	0	0	70	21
	500	100	40	60	20	100	-	90	0	40	80	
a-182	125	60	20	80	0	100	60	50	0	0	50	21
	500	60	60	80	10	100	-	50	0	20	90	
a-183	500	0	40	10	10	100	100	0	0	10	40	21
a-184	500	50	20	10	20	100	-	0	0	0	40	21
a-187	500	70	20	100	40	95	-	50	0	0	60	21
a-188	500	50	10	90	40	95	-	10	0	0	70	21
a-189	500	90	10	100	40	95	-	90	0	0	70	21
a-191	125	90	30	90	20	90	-	20	0	0	50	21
	500	100	50	90	40	90	-	40	0	0	50	

Table 5 (continued)

Comp. No.	Dose of active ingredient (g/ha)	Growth-controlling degree (%)										Evaluation day
		EC	DS	AR	SS	PP	XS	OS	TR	ZM	GM	
a-192	500	30	0	30	0	80	90	30	0	10	60	21
a-194	500	0	30	20	0	100	90	0	0	0	50	22
a-200	500	70	50	95	20	100	90	20	0	0	40	26
a-201	500	20	40	90	40	20	70	40	0	0	0	26
a-203	125 500	20 70	60 80	90 100	10 50	80 100	80 95	20 90	0 0	0 0	60 90	22
a-204	500	70	50	30	0	80	80	20	0	0	50	22
a-206	125 500	80 100	30 80	30 100	20 50	100 100	100 100	90 100	0 0	0 70	90 100	22
a-207	125 500	70 90	20 60	20 90	0 40	100 100	80 100	80 100	0 0	0 20	80 100	22
a-208	125 500	30 70	40 60	90 70	10 40	80 90	100 100	50 95	0 0	0 0	50 60	22
a-209	500	40	10	30	0	90	80	70	0	0	70	22
a-210	125 500	90 100	60 90	70 100	10 40	90 100	90 90	90 90	0 0	0 60	80 100	27
a-211	125 500	40 80	20 50	70 90	0 20	50 70	80 90	70 70	0 0	0 0	40 50	27
a-212	125 500	70 100	0 10	40 70	0 0	0 90	70 80	60 80	0 0	0 0	50 80	27
a-213	125 500	100 100	80 100	95 100	30 70	100 100	100 100	80 100	0 0	20 70	95 100	26
a-214	125 500	60 100	60 90	100 100	20 70	90 100	70 90	100 100	0 0	0 0	50 70	27
a-215	500	0	0	90	20	90	80	0	0	0	40	23

Table 5 (continued)

Comp. No.	Dose of active ingredient (g/ha)	Growth-controlling degree (%)										Evaluation day
		EC	DS	AR	SS	PP	XS	OS	TR	ZM	GM	
a-216	125	50	70	90	10	70	70	50	0	0	40	23
	500	90	90	100	60	80	100	100	0	0	70	
a-217	125	60	30	50	10	100	80	70	0	0	70	20
	500	30	60	80	20	100	100	100	0	70	100	
a-218	125	70	20	10	10	90	70	70	0	0	80	20
	500	90	60	50	20	100	80	90	0	40	100	
a-219	125	70	30	10	0	100	80	50	0	0	80	20
	500	100	70	60	30	100	100	80	0	70	100	
a-220	125	10	10	10	0	70	80	10	0	0	40	20
	500	70	30	30	20	90	80	60	0	0	40	
a-221	125	60	0	0	0	80	80	50	0	0	70	20
	500	80	60	40	30	100	100	50	0	0	90	
a-222	125	30	10	30	10	80	70	20	0	0	40	20
	500	80	50	80	50	90	90	70	0	10	60	
a-257	125	70	10	10	0	90	70	20	0	0	50	28
	500	90	60	10	10	100	100	30	0	30	100	
a-258	125	50	10	30	0	90	70	10	0	0	40	28
	500	90	70	80	30	100	100	50	0	0	70	
a-259	125	70	70	0	0	80	90	10	0	0	60	28
	500	90	100	20	0	90	100	40	0	0	80	
b-1	500	80	50	80	20	90	-	60	-	20	60	19
b-2	500	90	20	70	20	90	60	60	-	0	60	18

TEST EXAMPLE 2

Paddy field soil was put into a 1/1,000,000ha pot, and seeds of barnyardgrass (Echinochloa crus-galli L.) and japanese bulrush (Scirpus juncoides) were sown and slightly covered with soil. Then, the pot was left to stand still in a greenhouse in a state where the depth of flooding water was from 0.5 to 1 cm, and two days later, tubers of japanese ribbon wapato (Sagittaria pygmaea) were planted. Thereafter, the depth of flooding water was maintained at a level of from 3 to 4 cm, and when barnyardgrass and japanese bulrush reached a 0.5 leaf stage and japanese ribbon wapato reached to a primary leaf stage, an aqueous diluted solution of a wettable powder having the compound of the present invention formulated in accordance with a usual formulation method, was uniformly applied under submerged condition by a pipette so that the dose of the active ingredient would be at a predetermined level.

On the other hand, paddy field soil was put into a 1/1,000,000ha pot and puddled and leveled, and the depth of flooding water was from 3 to 4 cm. One day later, rice (Oryza sativa L. var. Nihonbare) of 2 leaf stage was transplanted in a depth of 3 cm. On the 4th day after the transplantation, the compound of the present invention was applied in the same manner as described above.

On the 14th days after the application of the

herbicide, the growth of barnyard grass, japanese burlrush and japanese ribbon wapato was visually observed and on the 21st day after the application the herbicide, the growth of rice was visually observed, and the herbicidal effects were evaluated by the growth-controlling degrees (%) ranging from 0 (equivalent to the non-treated area) to 100 (complete kill), whereby the results shown in Table 6 were obtained. Compound Nos. in Table 6 correspond to Compound Nos. in Table 4a and 4b given hereinbefore. The growth controlling degrees against rice of compounds Nos. a-101 et seq (except for a-131, a-132, a-145, a-146 and b-1) are mean values of two test results.

Table 6

Comp. No.	Dose of active ingredient (g/ha)	Growth-controlling degree (%)			
		EC	SJ	SP	OS
a-1	1000	50	100	100	—
	500	40	100	—	—
a-2	500	90	95	90	0
	250	85	85	85	0
a-3	1000	95	100	95	10
	500	95	100	85	0
a-4	500	100	85	85	0
	250	100	50	60	0
a-5	1000	40	85	85	90
	500	0	85	70	30
a-6	500	100	90	90	50
	250	100	85	85	30
a-7	500	100	100	85	80
	250	99	95	85	70
a-8	1000	0	85	85	0
	500	0	50	85	0
a-9	500	10	50	70	30
a-10	500	80	50	85	0
	250	60	50	85	0
a-11	250	100	95	90	100
	125	100	90	85	100
a-12	500	100	100	90	100
	250	100	100	90	100
a-13	500	100	100	90	100
	250	100	100	90	100
a-14	500	40	85	90	10
	250	10	85	85	0
a-15	500	100	85	70	0
	250	50	70	70	0
a-16	500	100	50	30	30
	250	99	0	30	0
a-17	500	100	85	50	20
	250	99	50	50	0

Table 6 (continued)

Comp. No.	Dose of active ingredient (g/ha)	Growth-controlling degree (%)			
		EC	SJ	SP	OS
a-18	500 250	0 0	90 85	95 95	0 0
a-19	500 250	70 40	90 90	90 85	10 0
a-20	500 250	90 85	85 50	30 0	60 35
a-21	500 250	50 30	95 90	90 50	30 30
a-22	500 250	95 100	95 95	90 50	30 20
a-23	500 250	100 50	99 99	90 50	0 0
a-24	250 125	40 20	85 80	90 85	10 0
a-25	250 125	100 85	90 85	90 85	0 0
a-26	1000 500	90 50	90 90	10 10	10 0
a-27	250 125	100 100	95 90	90 85	100 40
a-39	125 63	100 100	90 85	85 50	35 30
a-40	125 63	100 100	100 99	90 90	80 40
a-41	125 63	100 100	95 90	85 85	100 70
a-42	250 125	100 100	90 90	90 0	90 90
a-43	250 125	100 100	90 85	90 50	90 99
a-44	250 125	100 100	95 90	90 85	95 80
a-45	250 125	100 100	99 90	85 50	100 90

Table 6 (continued)

Comp. No.	Dose of active ingredient (g/ha)	Growth-controlling degree (%)			
		EC	SJ	SP	OS
a-47	250 125	100 100	100 85	95 60	99 70
a-48	250 125	100 99	95 85	90 50	95 50
a-49	250 125	100 100	90 90	85 50	100 100
a-51	250 125	100 100	99 95	85 10	90 60
a-52	250 125	100 100	99 90	30 30	100 95
a-53	125 63	100 100	95 90	90 90	80 0
a-55	250 125	100 100	95 50	20 0	30 30
a-58	250 125	100 99	85 70	50 50	40 40
a-59	63 31	100 100	90 50	10 -	30 30
a-60	250 125	100 100	100 95	70 30	95 70
a-69	250 125	100 100	99 85	50 50	100 35
a-72	125 63	90 60	85 85	20 20	40 35
a-73	125 63	100 100	90 70	50 50	80 30
a-76	125 63	100 100	90 85	30 0	0 0
a-77	125 63	90 70	85 50	50 30	70 40
a-78	250 125	100 85	85 60	85 85	80 30
a-79	250 125	100 100	95 85	90 50	90 40

Table 6 (continued)

Comp. No.	Dose of active ingredient (g/ha)	Growth-controlling degree (%)			
		EC	SJ	SP	OS
a-80	500 250	100 99	100 100	95 95	90 90
a-81	500 250	95 85	99 90	90 50	30 30
a-82	125 63	40 20	85 10	50 0	35 20
a-83	250 125	99 95	99 99	90 90	60 40
a-84	250 125	100 100	99 95	50 50	90 50
a-85	250 125	100 95	85 70	50 0	20 20
a-86	125 63	10 10	70 60	10 10	0 0
a-87	250 125	100 100	90 85	30 0	40 30
a-88	250 125	100 100	90 90	50 20	100 95
a-89	250 125	100 99	99 99	90 50	99 95
a-90	250 125	99 99	95 90	90 85	80 70
a-91	500 250	99 99	99 95	90 85	35 30
a-92	250 125	99 85	90 85	10 0	35 10
a-93	250 125	100 100	100 100	— 85	40 0
a-94	250 125	100 100	99 95	90 10	99 99
a-95	250 125	100 100	100 100	— 85	— —
a-96	250 125	100 100	95 90	85 10	100 99

Table 6 (continued)

Comp. No.	Dose of active ingredient (g/ha)	Growth-controlling degree (%)			
		EC	SJ	SP	OS
a-97	63 31	95 95	30 0	50 0	50 50
a-98	63 31	100 100	80 60	70 0	60 30
a-99	125 63	100 100	95 —	50 50	100 100
a-100	125 63	50 10	70 10	10 0	30 0
a-101	63 31	100 80	10 0	0 0	40 30
a-102	63 31	95 85	85 50	50 0	40 25
a-103	63 31	95 85	90 10	50 30	65 10
a-104	63 31	99 99	0 0	0 0	30 30
a-105	63 31	95 90	0 0	0 0	40 10
a-106	250 125	100 99	70 30	70 50	50 15
a-118	250 125	100 100	90 70	60 60	95 55
a-120	250 125	100 100	99 90	50 40	98 90
a-122	250 125	100 100	100 100	70 60	95 100
a-127	1000	20	60	70	—
a-131	250 125	100 100	90 85	— 90	80 50
a-132	250 125	100 100	85 50	— 50	— 30
a-135	250 125	100 70	20 0	0 0	10 0
a-138	125 63	100 80	20 —	0 0	5 15

Table 6 (continued)

Comp. No.	Dose of active ingredient (g/ha)	Growth-controlling degree (%)			
		EC	SJ	SP	OS
a-139	500 250	70 70	60 20	50 20	— —
a-140	250 125	100 100	90 90	30 —	98 35
a-141	250 125	100 100	100 100	30 0	100 95
a-143	500 250	100 100	10 0	30 10	— —
a-144	500 250	99 70	20 10	90 85	— —
a-145	250 125	100 100	60 50	50 40	0 10
a-146	500 250	90 60	90 70	90 70	30 0
a-148	125 63	100 100	70 70	0 0	0 0
a-149	125 63	90 85	30 10	0 0	0 0
a-150	250 125	100 95	50 0	60 30	0 5
a-151	250 125	100 100	0 0	0 —	30 5
a-152	250 125	100 100	60 30	0 0	100 100
a-153	250 125	99 100	90 30	70 30	10 20
a-154	250 125	100 99	20 0	40 0	60 20
a-155	250 125	100 100	70 60	0 0	85 50
a-157	250 125	90 90	95 0	70 0	0 0
a-158	250 125	100 100	80 50	30 60	90 90

Table 6 (continued)

Comp. No.	Dose of active ingredient (g/ha)	Growth-controlling degree (%)			
		EC	SJ	SP	OS
a-160	250 125	80 70	50 —	0 0	0 0
a-161	250 125	70 70	0 0	0 0	5 0
a-162	250 125	100 100	0 0	0 0	10 0
a-164	250 125	100 100	0 0	0 0	60 10
a-165	125 63	100 95	80 60	30 0	100 35
a-166	125 63	100 100	60 20	0 0	100 85
a-167	250 125	100 100	95 70	60 0	35 10
a-168	125 63	100 100	95 90	0 0	55 50
a-169	125 63	100 100	40 40	0 0	90 55
a-170	125 63	100 100	30 10	0 0	85 45
a-171	250	70	0	0	0
a-172	250	80	30	50	50
a-173	250 125	99 70	20 —	0 0	100 25
a-174	250 125	100 100	60 60	0 0	100 100
a-175	250 125	80 80	50 30	0 0	0 0
a-177	250 125	100 100	50 30	0 0	0 0
a-178	250 125	100 100	30 30	0 0	10 0
a-179	250 125	100 100	90 30	0 0	100 90

Table 6 (continued)

Comp. No.	Dose of active ingredient (g/ha)	Growth-controlling degree (%)			
		EC	SJ	SP	OS
a-180	250 125	100 100	30 0	0 0	80 60
a-181	250	100	0	20	50
a-182	250 125	100 100	70 0	0 0	45 15
a-184	250	85	0	0	0
a-185	250	70	0	0	0
a-187	250 125	70 30	80 80	20 0	15 0
a-188	250 125	100 100	50 50	20 0	95 80
a-189	250 125	100 100	50 50	20 0	55 50
a-191	250 125	100 95	20 0	20 0	35 0
a-194	250	90	30	0	10
a-200	250 125	100 100	90 90	0 0	70 80
a-202	250 125	100 85	70 20	0 0	10 10
a-203	250 125	100 100	100 95	95 95	100 100
a-204	250 125	100 99	30 0	30 0	10 5
a-205	250	85	0	0	5
a-206	250 125	100 95	0 0	60 70	100 90
a-207	250 125	95 80	0 0	60 80	50 25
a-208	250 125	100 100	30 0	70 60	80 50
a-210	250 125	100 100	40 70	70 60	100 85

Table 6 (continued)

Comp. No.	Dose of active ingredient (g/ha)	Growth-controlling degree (%)			
		EC	SJ	SP	OS
a-213	250 125	100 100	50 20	0 0	95 65
a-214	250 125	100 99	60 60	20 0	60 15
a-216	250 125	100 100	80 40	0 0	95 15
a-217	250 125	100 95	20 0	0 0	100 50
a-218	250 125	100 70	70 30	20 0	100 85
a-219	250 125	100 100	30 0	0 0	100 70
a-230	63 31	100 100	0 0	0 0	— —
a-232	63 31	100 100	30 0	— 0	— —
a-233	63 31	70 50	0 0	0 0	— —
a-239	63 31	100 80	0 0	0 0	0 0
a-249	250 125	100 100	0 0	0 0	95 40
a-250	63 31	100 100	40 0	0 0	80 45
a-253	63 31	100 100	0 0	0 0	35 30
a-254	63 31	100 100	0 0	— 0	5 0
a-256	63 31	100 80	0 0	0 0	0 0
a-257	250 125	100 100	20 0	0 0	0 0
a-258	250 125	60 70	0 0	20 0	5 0

Table 6 (continued)

Comp. No.	Dose of active ingredient (g/ha)	Growth-controlling degree (%)			
		EC	SJ	SP	OS
a-259	250 125	100 100	0 0	0 0	10 0
a-260	250 125	100 85	0 0	20 0	10 0
a-261	250 125	100 70	0 0	40 40	5 0
a-262	250 125	100 100	10 0	0 0	10 0
a-263	250 125	100 95	0 0	0 0	10 0
a-264	250 125	85 70	0 0	0 0	30 15
a-271	63 31	100 95	0 0	0 0	— —
b-1	250 125	100 100	85 50	50 50	50 20

Now, Formulation Examples of the present invention will be given. Compound Nos. in Formulation Examples correspond to Compound Nos. in Table 4a to 4b given hereinbefore.

FORMULATION EXAMPLE 1

(1) Compound No. a-12	75 parts by weight
(2) Sodium N-methyl-N-oleoyl taurate (Geropon T-77, tradename, manufactured by Rhone-Poulenc)	14.5 parts by weight
(3) NaCℓ	10 parts by weight
(4) Dextrin	0.5 part by weight

The above components are placed in a high-speed mixing granulator, admixed with 20 wt% of water, granulated, and dried to form water-dispersible granules.

FORMULATION EXAMPLE 2

(1) Kaolin	78 parts by weight
(2) Condensate of sodium naphthalene sulfonate and formalin (Laveline FAN, tradename, manufactured by Dai-ichi Kogyo Seiyaku Co., Ltd.)	2 parts by weight
(3) Sodium polyoxyethylene alkylaryl ether sulfate-premix with white carbon (Sorpol 5039, tradename, manufactured by Toho Chemical Industry Co., Ltd.)	5 parts by weight
(4) White carbon (Carplex, tradename, manufactured by Shionogi Seiyaku Co., Ltd.)	15 parts by weight

The mixture of the above components (1) to (4) and Compound No. a-6 are mixed in a weight ratio of 9:1 to obtain a wettable powder.

FORMULATION EXAMPLE 3

(1)	Talc micropowder (Hi-Filler No. 10, tradename, manufactured by Matsumura Sangyo Co., Ltd.)	33 parts by weight
(2)	Dialkyl sulfosuccinate-premixed with white carbon (Sorpol 5050, tradename, manufactured by Toho Chemical Industry Co., Ltd.)	3 parts by weight
(3)	A mixture of polyoxyethylene alkylaryl ether sulfate and a polyoxyethylene monomethyl ether carbonate, premixed with white carbon (Sorpol 5073, tradename, manufactured by Toho Chemical Industry Co., Ltd.)	4 parts by weight
(4)	Compound No. a-42	60 parts by weight

The above components (1) to (4) are mixed to obtain a wettable powder.

FORMULATION EXAMPLE 4

(1)	Compound No. a-27	4 parts by weight
(2)	Bentonite	30 parts by weight
(3)	Calcium carbonate	61.5 parts by weight
(4)	Polycarboxylic acid type surfactant (Toxanon GR-31A, tradename, manufactured by Sanyo Chemical Industries Co., Ltd.)	3parts by weight
(5)	Calcium lignin sulfonate	1.5 parts by weight

Pulverized component (1) and components (2) and (3) are preliminarily mixed, and then components (4) and (5) and water are mixed thereto. The mixture is extruded and granulated, followed by drying and size-adjusting to obtain granules.

FORMULATION EXAMPLE 5

(1)	Compound No. a-22	30 parts by weight
(2)	A pulverized product of a mixture of kaolinite and sericite (Zieclite, tradename, manufactured by Zieclite Co., Ltd.)	60 parts by weight
(3)	Alkyl naphthalene sulfonate (New Kalgen WG-1, tradename, manufactured by Takemoto Oils and Fats Co., Ltd.)	5 parts by weight
(4)	Polyoxyalkylene allyl phenyl ether sulfate (New Kalgen FS-7, tradename, manufactured by Takemoto Oils and Fats Co., Ltd.)	5 parts by weight

Components (1), (2) and (3) are mixed and passed through a pulverizer, and then component (4) is added thereto. The mixture is kneaded and then extruded and granulated, followed by drying and size-adjusting to obtain water-dispersible granules.

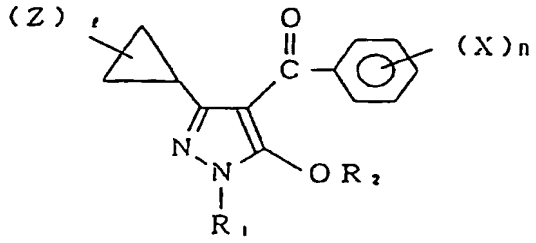
FORMULATION EXAMPLE 6

(1)	Compound No. a-13	28 parts by weight
(2)	Triethanolamine salts of oxyethylated polyarylphenol phosphate (Soprophor FL, tradename, manufactured by Rhone-Poulenc)	2 parts by weight
(3)	A mixture of polyoxyethylene styryl phenyl ether and alkyl aryl sulfonate (Sorpol 355, tradename, manufactured by Toho Chemical Industry Co., Ltd.)	1 part by weight
(4)	Isoparaffin hydrocarbon (IP solvent 1620, tradename, manufactured by Idemitsu Petrochemical Co., Ltd.)	32 parts by weight
(5)	Ethylene glycol	6 parts by weight
(6)	Water	31 parts by weight

The above components (1) to (6) are mixed and pulverized by a wet-grinding machine (Dyno-mill) to obtain a water-based suspension concentrate.

CLAIMS

1. A pyrazole compound of the formula (I) or its salt:

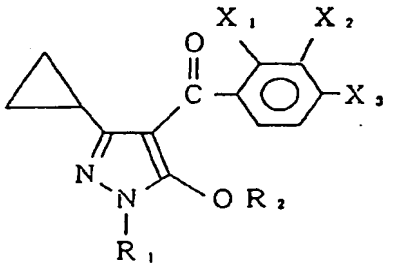


(I)

wherein R_1 is an alkyl group, R_2 is a hydrogen atom, a methyl group, $-A-R_3$, a phenyl group which may be substituted, a pyridyl group which may be substituted, or an allyl group which is substituted by a phenyl group, A is $-SO_2-$, $-CO-$, $-CH(R_6)-$ or $-CH(R_7)CO-$, R_3 is an alkyl group which may be substituted, an alkenyl group which may be substituted, an alkynyl group which may be substituted, an alkoxy group which may be substituted, a cyano group, a dialkylamino group or a phenyl group which may be substituted, each of R_6 and R_7 is a hydrogen atom or an alkyl group, X is a hydrogen atom, a halogen atom, an alkyl group, a haloalkyl group, an alkoxy group, an alkylthio group, an alkylsulfinyl group, an alkylsulfonyl group, a nitro group, an alkoxycarbonyl group, $-SO_2N(R_8)R_9$, $-N(R_{10})SO_2R_{11}$, $-CH_2S(O)qR_{12}$ or $-OSO_2R_{13}$, each of R_8, R_9, R_{10}, R_{11}, R_{12} and R_{13} is an alkyl group, Z is an alkyl group, l is an integer of from 0 to 5, n is an integer of from 1 to 5, and q is an integer of from 0 to 2, provided that when l is at least 2, a plurality of Z

may be the same or different, and when n is at least 2, a plurality of X may be the same or different.

2. The pyrazole compound or its salt according to Claim 1, wherein the formula (I) is represented by the formula (I'):



(I')

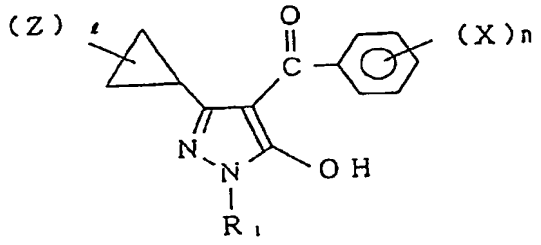
wherein R_1 is an alkyl group, R_2 is a hydrogen atom or $-A-R_3$, A is $-SO_2-$, $-CO-$, $-CH_2-$ or $-CH_2CO-$, R_3 is an alkyl group which may be substituted, an alkenyl group which may be substituted, an alkynyl group which may be substituted , a cyano group or a phenyl group which may be substituted, each of X^1, X^2 and X^3 is a hydrogen atom, a halogen atom, an alkyl group, a haloalkyl group, an alkoxy group, an alkylthio group, an alkylsulfinyl group, an alkylsulfonyl group, a nitro group, $-SO_2N(R_8)R_9$, $-N(R_{10})SO_2R_{11}$, $-CH_2S(O)qR_{12}$ or $-OSO_2R_{13}$, each of R_8, R_9, R_{10}, R_{11}, R_{12} and R_{13} is an alkyl group, and q is an integer of from 0 to 2.

3. The pyrazole compound or its salt according to Claim 2, wherein A is $-SO_2-$, $-CH_2-$ or $-CH_2CO-$, each of X^1, X^2 and X^3 is a hydrogen atom, a halogen atom, an alkyl group, a haloalkyl group, an alkoxy group, an alkylthio

group, an alkylsulfinyl group, an alkylsulfonyl group or a nitro group.

4. The pyrazole compound or its salt according to Claim 3, wherein X^1 is an alkylthio group, an alkylsulfinyl group or an alkylsulfonyl group, and each of X^2 and X^3 is a hydrogen atom, a halogen atom, an alkyl group, a haloalkyl group or a nitro group.

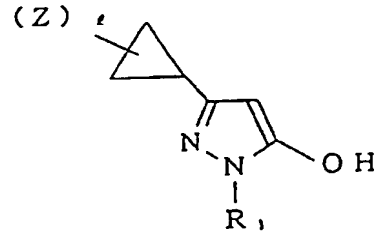
5. A process for producing a pyrazole compound of the formula (I-1) or its salt:



(I-1)

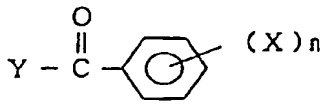
wherein R_1 is an alkyl group, Z is an alkyl group, l is an integer of from 0 to 5, provided that when l is at least 2, a plurality of Z may be the same or different, X is a hydrogen atom, a halogen atom, an alkyl group, a haloalkyl group, an alkoxy group, an alkylthio group, an alkylsulfinyl group, an alkylsulfonyl group, a nitro group, an alkoxycarbonyl group, $-SO_2N(R_8)R_9$, $-N(R_{10})SO_2R_{11}$, $-CH_2S(O)qR_{12}$ or $-OSO_2R_{13}$, each of R_8, R_9, R_{10}, R_{11}, R_{12} and R_{13} is an alkyl group, q is an integer of from 0 to 2, and n is an integer of from 1 to 5, provided that when n is at least 2, a plurality of X may be the same or different, which comprises reacting a

compound of the formula (II):



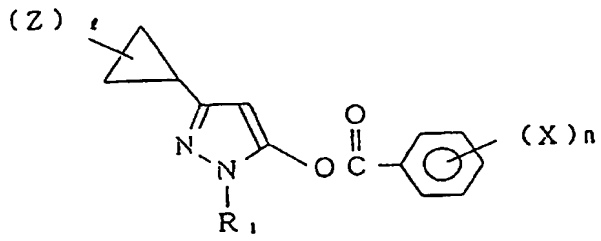
(II)

wherein R_1, Z and l are as defined above, with a compound of the formula (III):



(III)

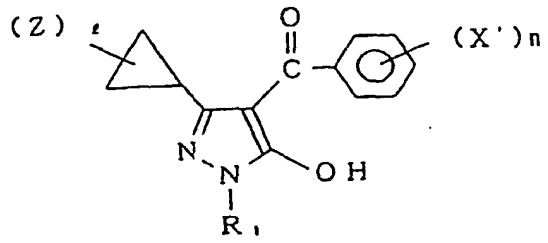
wherein X and n are as defined above, and Y is a halogen atom, to obtain a compound of the formula (IV):



(IV)

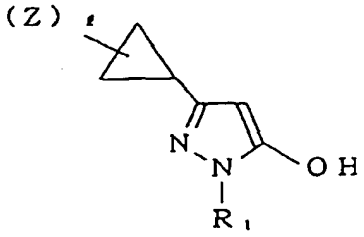
wherein R_1, X, Z, l and n are as defined above, and subjecting the compound of the formula (IV) to a rearrangement reaction.

6. A process for producing a pyrazole compound of the formula (I-l') or its salt:



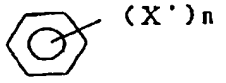
(I-1')

wherein R_1 is an alkyl group, Z is an alkyl group, l is an integer of from 0 to 5, provided that when l is at least 2, a plurality of Z may be the same or different, X' is a hydrogen atom, a halogen atom, an alkyl group, a haloalkyl group, an alkoxy group, an alkylthio group, an alkylsulfinyl group or an alkylsulfonyl group, and n is an integer of from 1 to 5, provided that when n is at least 2, a plurality of X' may be the same or different, which comprises reacting a compound of the formula (II):



(II)

wherein R_1, Z and l are as defined above, with a compound of the formula (V):

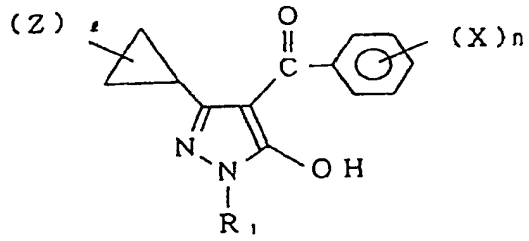


(V)

wherein X' and n are as defined above, and carbon tetrachloride, followed by a hydrolytic reaction.

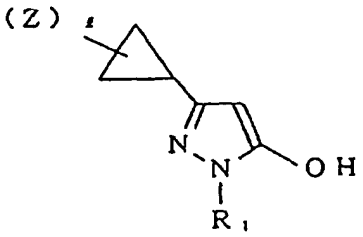
7. A process for producing a pyrazole compound of the

formula (I-1) or its salt:



(I-1)

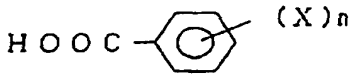
wherein R_1 is an alkyl group, Z is an alkyl group, l is an integer of from 0 to 5, provided that when l is at least 2, a plurality of Z may be the same or different, X is a hydrogen atom, a halogen atom, an alkyl group, a haloalkyl group, an alkoxy group, an alkylthio group, an alkylsulfinyl group, an alkylsulfonyl group, a nitro group, an alkoxycarbonyl group, $-SO_2N(R_8)R_9$, $-N(R_{10})SO_2R_{11}$, $-CH_2S(O)qR_{12}$ or $-OSO_2R_{13}$, each of R_8, R_9, R_{10}, R_{11}, R_{12} and R_{13} is an alkyl group, q is an integer of from 0 to 2, and n is an integer of from 1 to 5, provided that when n is at least 2, a plurality of X may be the same or different, which comprises reacting a compound of the formula (II):



(II)

wherein R_1, Z and l are as defined above, with a compound

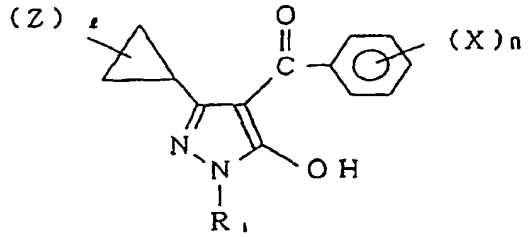
of the formula (VI):



(VI)

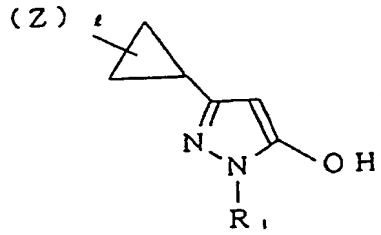
wherein X and n are as defined above.

8. A process for producing a pyrazole compound of the formula (I-1) or its salt:



(I-1)

wherein R_1 is an alkyl group, Z is an alkyl group, l is an integer of from 0 to 5, provided that when l is at least 2, a plurality of Z may be the same or different, X is a hydrogen atom, a halogen atom, an alkyl group, a haloalkyl group, an alkoxy group, an alkylthio group, an alkylsulfinyl group, an alkylsulfonyl group, a nitro group, an alkoxycarbonyl group, $-SO_2N(R_8)R_9$, $-N(R_{10})SO_2R_{11}$, $-CH_2S(O)_qR_{12}$ or $-OSO_2R_{13}$, each of R_8, R_9, R_{10}, R_{11}, R_{12} and R_{13} is an alkyl group, q is an integer of from 0 to 2, and n is an integer of from 1 to 5, provided that when n is at least 2, a plurality of X may be the same or different, which comprises reacting a compound of the formula (II):



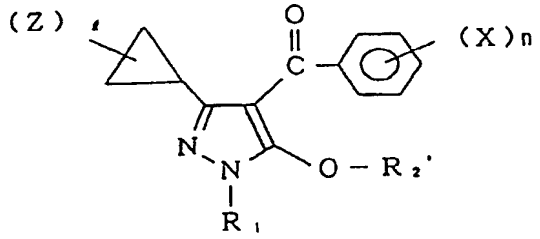
(II)

wherein R_1, Z and l are as defined above, with a compound of the formula (X):

(X)

wherein X and n are as defined above, and T is a chlorine atom, a bromine atom or an iodine atom, and carbon monoxide.

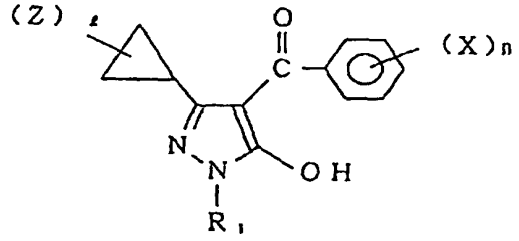
9. A process for producing a pyrazole compound of the formula (I-2) or its salt:



(I-2)

wherein R_1 is an alkyl group, R_2' is a methyl group, $-A-R_3$, a phenyl group which may be substituted, a pyridyl group which may be substituted or an allyl group which is substituted by a phenyl group, A is $-SO_2-$, $-CO-$, $-CH(R_6)-$ or $-CH(R_7)CO-$, R_3 is an alkyl group which may be

substituted, an alkenyl group which may be substituted, an alkynyl group which may be substituted, an alkoxy group which may be substituted, a cyano group, a dialkylamino group or a phenyl group which may be substituted, each of R_6 and R_7 is a hydrogen atom or an alkyl group, Z is an alkyl group, l is an integer of from 0 to 5, provided that when l is at least 2, a plurality of Z may be the same or different, X is a hydrogen atom, a halogen atom, an alkyl group, a haloalkyl group, an alkoxy group, an alkylthio group, an alkylsulfinyl group, an alkylsulfonyl group, a nitro group, an alkoxycarbonyl group, $-SO_2N(R_8)R_9$, $-N(R_{10})SO_2R_{11}$, $-CH_2S(O)_qR_{12}$ or $-OSO_2R_{13}$, each of R_8, R_9, R_{10}, R_{11}, R_{12} and R_{13} is an alkyl group and n is an integer of from 1 to 5, provided that when n is at least 2, a plurality of X may be the same or different, q is an integer of from 0 to 2, which comprises reacting a compound of the formula (I-1):



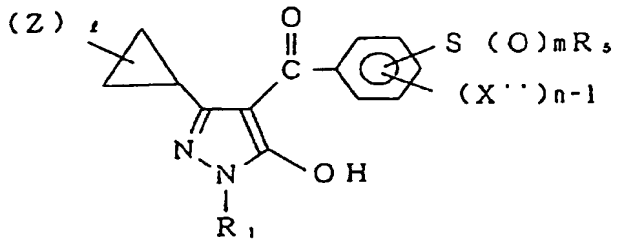
(I-1)

where R_1, X, Z, n and l are as defined above, with a compound of the formula (VII):

$$Y\text{-}R_2' \qquad \text{(VII)}$$

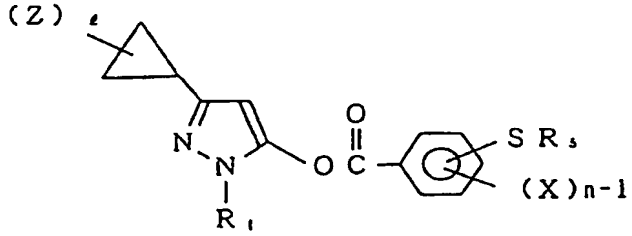
wherein R_2' is as defined above, and Y is a halogen atom.

10. A process for producing a pyrazole compound of the formula (I-4) or its salt:



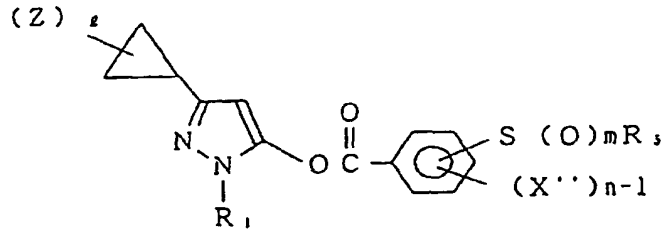
(I-4)

wherein each of R_1 and R_5 is an alkyl group, Z is an alkyl group, l is an integer of from 0 to 5, provided that when l is at least 2, a plurality of Z may be the same or different, X" is a hydrogen atom, a halogen atom, an alkyl group, a haloalkyl group, an alkoxy group, an alkylsulfinyl group, an alkylsulfonyl group, a nitro group, an alkoxycarbonyl group, $-SO_2N(R_8)R_9$, $-N(R_{10})SO_2R_{11}$, $-CH_2S(O)_{q'}R_{12}$ or $-OSO_2R_{13}$, each of R_8, R_9, R_{10}, R_{11}, R_{12} and R_{13} is an alkyl group, q' is 1 or 2, m is 1 or 2, and n is an integer of from 1 to 5, provided that when n is at least 3, a plurality of X" may be the same or different, which comprises oxidizing a compound of the formula (VI-1):



(VI-1)

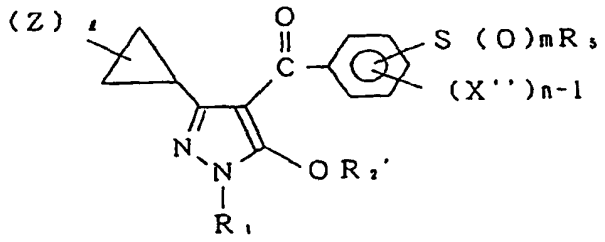
where R_1, R_5, Z, l and n are as defined above and X is a hydrogen atom, a halogen atom, an alkyl group, a haloalkyl group, an alkoxy group, an alkylthio group, an alkylsulfinyl group, an alkylsulfonyl group, a nitro group, an alkoxycarbonyl group, $-SO_2N(R_8)R_9$, $-N(R_{10})SO_2R_{11}$, $-CH_2S(O)_qR_{12}$ or $-OSO_2R_{13}$, each of R_8, R_9, R_{10}, R_{11}, R_{12} and R_{13} is an alkyl group, provided that when n is at least 3, a plurality of X may be the same or different to obtain a compound of the formula (VI-2):



(VI-2)

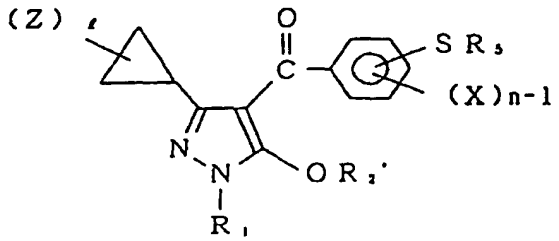
wherein R_1, R_5, Z, X", l, m and n are as defined above, and subjecting the compound of the formula (VI-2) to a rearrangement reaction.

11. A process for producing a pyrazole compound of the formula (I-7) or its salt:



(I-7)

wherein each of R_1 and R_5 is an alkyl group, R_2' is a methyl group, $-A-R_3$, a phenyl group which may be substituted, a pyridyl group which may be substituted or an allyl group which is substituted by a phenyl group, A is $-SO_2-$, $-CO-$, $-CH(R_6)-$ or $-CH(R_7)CO-$, R_3 is an alkyl group which may be substituted, an alkenyl group which may be substituted, an alkynyl group which may be substituted, an alkoxy group which may be substituted, a cyano group, a dialkylamino group or a phenyl group which may be substituted, each of R_6 and R_7 is a hydrogen atom or an alkyl group, Z is an alkyl group, l is an integer of from 0 to 5, provided that when l is at least 2, a plurality of Z may be the same or different, m is 1 or 2, X" is a hydrogen atom, a halogen atom, an alkyl group, a haloalkyl group, an alkoxy group, an alkylsulfinyl group, an alkylsulfonyl group, a nitro group, an alkoxycarbonyl group, $-SO_2N(R_8)R_9$, $-N(R_{10})SO_2R_{11}$, $-CH_2S(O)_{q'}R_{12}$ or $-OSO_2R_{13}$, each of R_8, R_9, R_{10}, R_{11}, R_{12} and R_{13} is an alkyl group, q' is 1 or 2, and n is an integer of from 1 to 5, provided that when n is at least 3, a plurality of X" may be the same or different, which comprises oxidizing a compound of the formula (I-6):



(I-6)

where R_1, R_2', R_5, Z, l and n are as defined above, and X is a hydrogen atom, a halogen atom, an alkyl group, a haloalkyl group, an alkoxy group, an alkylthio group, an alkylsulfinyl group, an alkylsulfonyl group, a nitro group, an alkoxycarbonyl group, $-SO_2N(R_8)R_9$, $-N(R_{10})SO_2R_{11}$, $-CH_2S(O)_qR_{12}$ or $-OSO_2R_{13}$, each of R_8, R_9, R_{10}, R_{11}, R_{12} and R_{13} is an alkyl group, q is an integer of from 0 to 2, provided that when n is at least 3, a plurality of X may be the same or different.

12. A herbicide containing the pyrazole compound or its salt as defined in Claim 1, as an active ingredient.

13. A method for controlling noxious weeds, which comprises applying an effective amount of the pyrazole compound or its salt as defined in Claim 1.

14. A method for controlling noxious weeds, which comprises applying an effective amount of the pyrazole compound or its salt as defined in Claim 1 to an upland field.

15. A method for controlling noxious weeds, which comprises applying an effective amount of the pyrazole compound or its salt as defined in Claim 1 to a corn field.

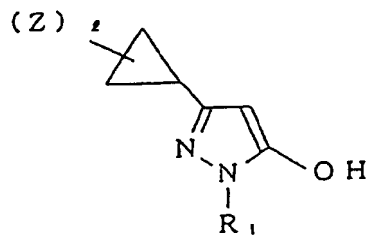
16. A method for controlling noxious weeds, which comprises applying an effective amount of the pyrazole compound or its salt as defined in Claim 1 to a wheat field.

17. A method for controlling noxious weeds, which

comprises applying an effective amount of the pyrazole compound or its salt as defined in Claim 1 to a paddy field.

18. A mixed herbicidal composition comprising at least one member selected from the pyrazole compound or its salt as defined in Claim 1 and at least one member selected from active ingredient compounds of other herbicides.

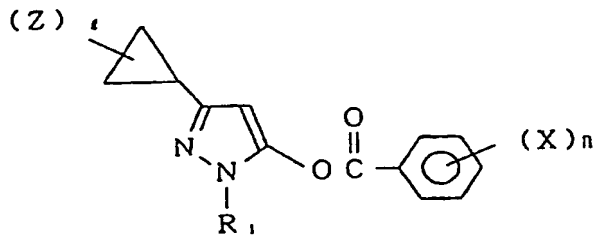
19. A compound of the formula (II):



(II)

wherein R_1 is an alkyl group, Z is an alkyl group and l is an integer of from 0 to 5, provided that when l is at least 2, a plurality of Z may be the same or different.

20. The compound of the formula (IV):



(IV)

wherein R_1 is an alkyl group, X is a hydrogen atom, a

halogen atom, an alkyl group, a haloalkyl group, an alkoxy group, an alkylthio group, an alkylsulfinyl group, an alkylsulfonyl group, a nitro group, an alkoxycabonyl group, $-SO_2N(R_8)R_9$, $-N(R_{10})SO_2R_{11}$, $-CH_2S(O)_qR_{12}$ or $-OSO_2R_{13}$, each of R_8, R_9, R_{10}, R_{11}, R_{12} and R_{13} is an alkyl group, Z is an alkyl group, l is an integer of from 0 to 5, n is an integer of from 1 to 5, and q is an integer of from 0 to 2, provided that when l is at least 2, a plurality of Z may be the same or different, and when n is at least 2, a plurality of X may be the same or different.

INTERNATIONAL SEARCH REPORT

Inten nal Application No
PCT/JP 97/01457

A. CLASSIFICATION OF SUBJECT MATTER
IPC 6 C07D231/20 C07D401/12 A01N43/56

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 6 C07D

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	CHEMICAL ABSTRACTS, vol. 114, no. 7, 18 February 1991 Columbus, Ohio, US; abstract no. 62091m, T. AONO ET AL.: "Preparation of pyrazolones and peroxylipid formation inhibitors, lipoxygenase inhibitors, and collagenase inhibitors containing them." page 690; column 2; XP002037186 see abstract; and Chemical Abstracts, CHEMICAL SUBSTANCES, 12th Collective Index, vol. 106-115, 1987-1991, page 78553CS, the compounds with the RN: [131645-10-8], [131645-56-2], and [131645-19-7] & JP 02 229 168 A (TAKEDA CHEMICAL INDUSTRIES, LTD.) 11 September 1990 --- -/--	19

[X] Further documents are listed in the continuation of box C.

[X] Patent family members are listed in annex.

* Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
7 August 1997	26. 08. 97

Name and mailing address of the ISA	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+ 31-70) 340-2040, Tx. 31 651 epo nl, Fax (+ 31-70) 340-3016	Fink, D

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INTERNATIONAL SEARCH REPORT

Inten nal Application No
PCT/JP 97/01457

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	US 4 063 925 A (KONOTSUNE TAKUO ET AL) 20 December 1977 see column 1, line 7 - column 2, line 10 see column 19; example 3 ---	1-4, 12-18
Y	EP 0 638 555 A (NISSAN CHEMICAL IND LTD) 15 February 1995 cited in the application see page 149 - page 150; claims ---	1-4, 12-18
A	GB 2 002 375 A (ISHIHARA MINING & CHEMICAL CO) 21 February 1979 cited in the application see the whole document -----	1-20

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INTERNATIONAL SEARCH REPORT

Information on patent family members

Inter. nal Application No
PCT/JP 97/01457

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
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